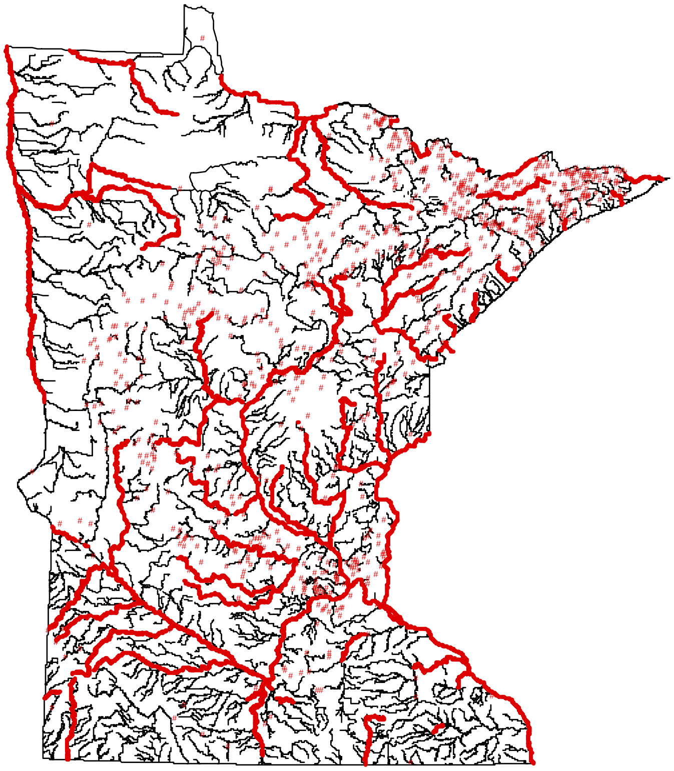

MINNESOTA STATEWIDE MERCURY TOTAL MAXIMUM DAILY LOAD

Final*

March 27, 2007

Minnesota Pollution Control Agency

*Approved by US EPA March 27, 2007

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Acknowledgements

The Minnesota Pollution Control Agency acknowledges the important contribution of the many stakeholders that have participated in Minnesota’s Mercury Reduction Initiative and the Impaired Waters Program. We appreciate the support and encouragement of the USEPA’s Region 5 and Headquarters staff. The main contributors to this mercury TMDL are the Mercury TMDL Project Technical Committee: Howard Markus, Bruce Monson, Edward Swain, Ned Brooks, and Anne Jackson.

Executive Summary

Mercury is a neurotoxin, meaning it damages the central nervous system. The developing nervous system is at the greatest risk for damage. Mercury is also a global pollutant; it is transmitted around the world and accumulates to levels in fish that are potentially toxic to humans and wildlife. This report sets a target for fish tissue concentration of mercury that is generally safe for human consumption, and translates the target to reduction goals for mercury sources.

Environmental contaminants are usually treated as media-specific—an air, water, or soil contaminant. Mercury is a multimedia pollutant: transported by air, stored in soil, and chemically transformed and bioaccumulated in water. Mercury reductions needed to achieve the target for safe fish consumption are translated to mercury emissions reductions, because 99 percent of mercury load to Minnesota's lakes and streams is from atmospheric deposition.

Because this report includes jargon from the Clean Water Act and the Clean Air Act, the following point of clarification is needed: this report will refer to water releases of mercury as "discharges" and air releases as "emissions." While stacks from air emission sources may be referred to as point sources in the air-permitting arena, the Total Maximum Daily Load (TMDL) concept arises from the Clean Water Act, where "point sources" refer to identifiable pipe conveyances and include wastewater and stormwater, which have National Pollutant Discharge Elimination System (NPDES) permits. Therefore, air sources (i.e., stacks) will be referred to as "point source emissions" and water sources (i.e., pipes) will be referred to as "point source discharges."

Impaired Waters List Section 303(d) of the Federal Clean Water Act requires every state to prepare a list of impaired waters. Minnesota's 2004 303(d) List ("Impaired Waters List") includes water quality impairments in 1892 lakes and river reaches. Two-thirds of those waters are impaired because of mercury (Figure ES- 1). The 1239 impairments by mercury consist of 820 lake impairments and 419 river impairments. Twelve lakes and 20 river reaches are impaired for mercury in fish tissue and in the water column; 808 lakes and 399 river reaches are impaired for fish tissue only.

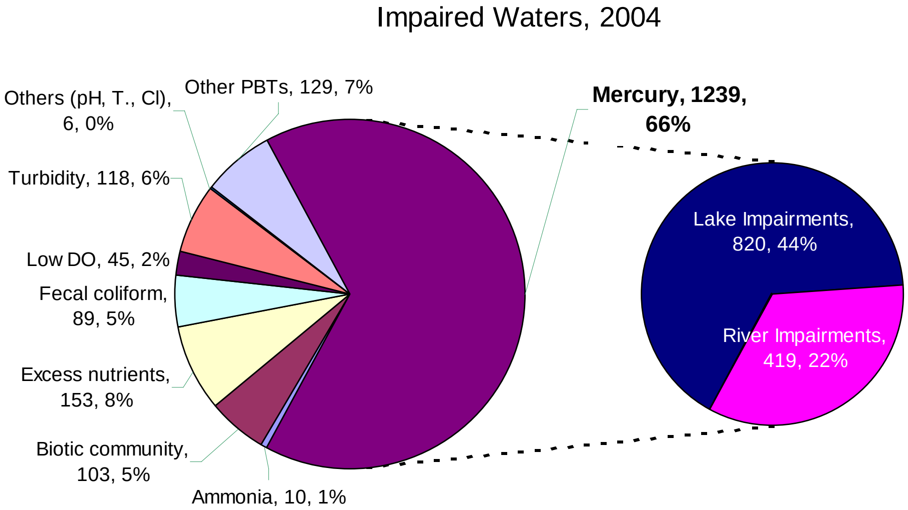

Figure ES- 1 Minnesota's 2004 Impaired Waters by Pollutant

Each impaired water is required to have a total maximum daily load study (TMDL). The TMDL is an evaluation of (1) pollutant sources, (2) pollutant load reduction needed to meet water quality standards and (3) allocation of the acceptable load to all sources. Because the source of essentially all mercury in Minnesota waters is atmospheric and, therefore, shared by all mercury-impaired waters of the state, the pollutant allocation to atmospheric sources will be the same for these waters. Seventy percent of atmospheric mercury deposition is from anthropogenic sources (i.e., from human activities) and the remaining thirty percent is from natural sources, such as volcanoes. Although state waters share common mercury sources, their capacity to assimilate the pollutant load varies because of differences in geography, water chemistry, and food webs. These differences are apparent in the geographic variation of mercury concentrations in fish, which is addressed through a regional approach.

Regional Approach The state is divided into two regional mercury TMDLs: northeast (NE) and a southwest (SW). The the two regions are separated by ecoregion boundaries (Figure ES- 2); NE comprises 41% of the state and SW covers 59%. Land-water mercury transport processes and concentrations in fish differ between the two regions. Land cover and geology controls transport processes and, consequently, water quality. NE region is dominated by forest and wetlands, and SW region is dominated by cultivated lands. Because there are similarities and differences between the two regions, the regional mercury TMDLs were developed (and described) in parallel.

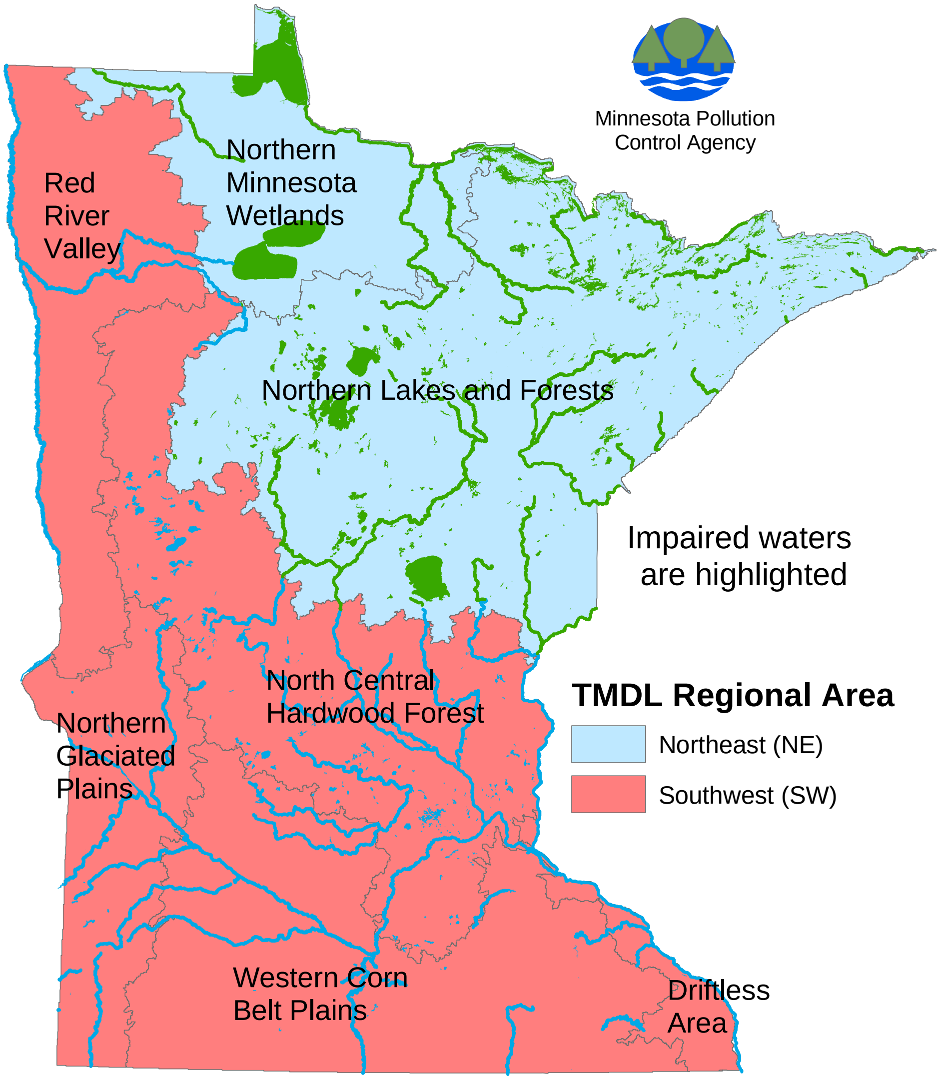

Figure ES- 2 TMDL Regional Areas and Mercury-Impaired Waters

TMDL Development The TMDL development follows a series of logical scientifically-based steps, beginning with establishment of the regional target level or endpoint goal (Table ES- 1). Both regions have the same fish tissue target level; however, because fish mercury concentrations differ by region, the

Table ES- 1 Summary of Minnesota's Regional Mercury TMDLs

This table summarizes the six steps to develop Minnesota's Mercury TMDL. See referenced report sections for more information about each step.		Units	Region	
			NE	SW
(1) The State is divided into two regions—Northeast (NE) and Southwest (SW)—based on differences in mercury's movement through the environment and fish tissue concentrations. [Section 4]	Regional Area in Minnesota	km^2	90,151	129,674
	Percent of total state area		41%	59%
(2) The TMDL target is the water quality criterion for mercury in fish. The 90th percentile fish tissue mercury concentration for a standard length walleye (40 cm) is compared to the fish tissue mercury criterion. The Reduction Factor is the percent reduction needed for fish to meet the water quality criterion. [Section 4.4]	**Target Level and Reduction Factor**			
	Target fish mercury concentration	mg/kg	0.2	0.2
	Mercury concentration for standard length walleye ($WE40_{90}$)	mg/kg	0.572	0.405
	Reduction Factor [$RF=(WE40_{90} - 0.2)/WE40_{90}$]		**65%**	**51%**
(3) Loads from mercury sources are summed by region for the 1990 mercury load that was either discharged from wastewater or deposited from air emissions. About 99% of the statewide mercury load was from nonpoint (air) sources in 1990. Nonpoint source load is the product of atmospheric deposition (12.5 grams per square kilometer per year) and regional area. Wastewater sources contribute about one percent of the total load in 1990 [Section 6.2][1]	**Mercury Load for Baseline Year – 1990**			
	Point Source Load (PSL; wastewater discharge)	kg/yr	26	7
	PSL percent of Total Source Load		2.2%	0.4%
	Nonpoint Source Load (NPSL; atmospheric deposition)	kg/yr	1127	1621
	Total Source Load (TSL)	kg/yr	**1153**	**1628**
(4) The Mercury TMDL is a loading goal equal to 1990 Total Source Load multiplied by (1- Reduction Factor), and is equal to wasteload allocation (WLA) plus load allocation (LA). The WLA consists of water point source discharges; it is set at one percent of the TMDL or equal to the estimated point source load, whichever is lower. The remainder of the TMDL is LA, which are atmospheric deposition sources. [Section 9][2]	**Final TMDL**			
	Mercury TMDL Loading Goal [TSL• (1– RF)]	kg/d	**1.10**	**2.18**
	Wasteload Allocation (WLA)	kg/d	**0.01**	**0.02**
	Load Allocation (LA)	kg/d	**1.09**	**2.16**
(5) To achieve the Mercury TMDL Goal, all load reductions must come from anthropogenic sources, which are 70% of the total atmospheric mercury deposition and are divided into in-state and out-of-state emission sources. In-state emission sources contribute 10% of the total mercury deposition, or 14.3% of the anthropogenic sources. The load is allocated to in-state and out-of-state contributions for both regions. The TMDL goal is converted to a mercury deposition flux (g km^{-2} yr^{-1}) when divided by the regional area. [Section 6.4]	**Mercury Load Allocation for In-state and Out-of-state Deposition Sources** [3]			
	In-State contribution to LA [0.143•LA)	kg/d	0.16	0.31
	Out-of-State contribution to LA [(1-0.143)•LA]	kg/d	0.94	1.86
	Mercury TMDL Atmospheric Deposition Goal	g km^{-2} yr^{-1}	4.4	6.1
	Necessary reduction from anthropogenic emission sources (RF ÷ 0.7)		**93%**	73%
(6) Because atmospheric mercury deposition is uniform across the entire state and in-state emissions disperse across both regions, the greater reduction goal, established for the northeast, becomes the statewide mercury load reduction goal, which is a 93% reduction in anthropogenic emissions. Subtracting the emissions reduction goal from the 1990 state emissions gives the state's TMDL mercury emissions goal. Since this TMDL uses 1990 as the starting point, it is informative to determine progress the state has made from 1990 to 2000. Between 1990 and 2005, 76% of the mercury emissions reduction goal was achieved, leaving 24% still to be met. [Section 6.4]	**Minnesota's Mercury TMDL Emissions Reduction Goal**		**Statewide**	
	State's mercury emissions for 1990	lb/yr	11,272	
	Emissions Reduction Goal (0.93 •1990 Emissions)	lb/yr	10,483	
	Minnesota's TMDL Mercury Emissions Goal (1990 Emissions – Reduction Goal)	**lb/yr**	**789**	
	Emissions reductions as of 2005 (70% of 1990 emissions)	lb/yr	7,931	
	Emissions reduction remaining as of 2005 to achieve goal	lb/yr	2,552	
	Percent of 1990 Emissions Reduction Goal remaining as of 2005		24%	

[1] For discussion of Reserve Capacity see Section 6.5

[2] For discussion of Margin of Safety see Section 7

[3] Minnesota's mercury deposition sources are 30% natural and 70% anthropogenic. The anthropogenic share is comprised of 30% global and 40% regional sources; one-fourth of the forty percent is from sources within Minnesota; therefore, the state's anthropogenic sources are 10% of total deposition (0.25 * 0.4). There are no significant natural sources (e.g., volcanoes and natural mercury mineral ores) in Minnesota. Mercury in Minnesota's soils is from atmospheric deposition.

necessary load reductions to achieve the goal differ by region. Minnesota's target level for mercury in fish is 0.2 mg/kg (parts per million, ppm), based on the EPA's development of a methylmercury criterion for fish tissue to protect human health. Minnesota's fish tissue mercury criterion is lower than EPA's 0.3 ppm criterion because of the higher fish consumption rate in the state. The 0.2 ppm corresponds to the Minnesota fish consumption advisory threshold for one meal per week—above that mercury concentration the consumption advice is one meal per month—for women who are pregnant or intending to become pregnant and children under 15 years of age.

Load reductions must be calculated relative to an appropriate baseline annual load. The most recent research that establishes total mercury deposition in Minnesota took place from 1988 to 1990. In addition, a baseline year of 1990 for this TMDL corresponds to the baseline year for Great Lakes mercury reduction goals and Minnesota's mercury emissions reduction goals. Prior to 1990 mercury use was relatively high and dropped precipitously beginning around 1990 as mercury was removed from many common products. Mercury deposition and mercury in fish tissue were probably in a relative steady state through 1990; therefore, comparing mercury deposition and fish tissue concentration in 1990 is most likely valid because of the steady state conditions leading up to 1990, but we have since entered a non-steady state period as mercury deposition declines.

For these regional TMDLs, target levels of mercury concentrations were determined in standard size top predator fish: northern pike (*Esox lucius*) and walleye (*Sanders vitreus*). Because mercury bioaccumulates and biomagnifies, concentration is highest at the top of the food web; therefore, achieving the mercury target concentration in the top predator fish will result in the whole food web, including the water column, achieving the target level. For the 1990 baseline year, fish tissue data were combined for each region for a five-year period—1988 to 1992—to account for annual weather fluctuations.

The target level of 0.2 ppm was applied to the 90th percentile mercury concentration. By protecting for the 90th percentile we expect to achieve the target level for other biota and for water concentrations of mercury. The difference between the regional 90th percentile concentration for the standard size fish and 0.2 ppm is the reduction factor (RF) needed to meet water quality standards. The RF is greater for the NE than the SW for both walleye and northern pike. Mercury concentrations in walleye were slightly higher than northern pike levels in both regions and, therefore, the RF for walleye was selected for load reduction calculations to provide a margin of safety. The resulting RFs are 65% for the NE and 51% for the SW.

The total source load (TSL) is the sum of the point source loads (PSL) and the non-point source loads (NPSL). Point source loads include the NPDES permitted facilities in the state, excluding cooling water discharges. PSL for the region is the product of facility design flow and the average measured effluent mercury for wastewater treatment plants in the state (5 ng Hg/L). Non-point source load is the product of atmospheric deposition flux in 1990 (12.5 g km^{-2} yr^{-1}) and regional surface area. The subsequent 1990 TSLs for NE and SW regions were 1153 kg/y and 1628 kg/y, respectively. About one percent of the TSL is attributable to PSL.

Mercury TMDL Goal for Minnesota Total mercury deposition in 1990 was 12.5 g km^{-2} yr^{-1} throughout the state. To achieve the target levels in fish tissue, the mercury deposition goals are 4.4 g km^{-2} yr^{-1} for the NE and 6.1 g km^{-2} yr^{-1} for the SW.

Mercury load reduction goals for each regional TMDL were calculated by applying the RF to the baseline mercury load. Reductions can only come from anthropogenic sources; therefore, load reduction goals require anthropogenic source reductions of 93% (65% reduction goal divided by 70% of total that is anthropogenic) in the NE region and 73% (51% of reduction goal divided by 70% anthropogenic) in the SW region.

Ten percent of the mercury deposition is attributed to anthropogenic sources within the state. Since natural sources cannot be controlled and are not expected to change, all mercury reductions must come from anthropogenic sources. The state's percentage of the anthropogenic sources is 14.3% (10% of total divided by 70% of total). The state's contributions to the load allocations (LA) are 0.16 kg/d for the NE and 0.31 kg/d for the SW. The out-of-state contributions to the LA are 0.94 kg/d for the NE and 1.86 kg/d for the SW.

Mercury Emission Reduction Goals Mercury load reduction goals are applied to emission reductions for the state. Atmospheric deposition of mercury is considered uniform across the state, and in-state emissions disperse across both regions; therefore, the emissions goal is applied statewide rather than by region. The northeast's greater regional reduction goal (i.e., 93% of anthropogenic sources) determines the TMDL's emission reduction goal. In 1990, the total mercury emissions from in-state sources were 11,272 lbs (5513 kg); the TMDL emissions goal is seven percent of the 1990 emissions: 789 lbs (358 kg). Minnesota's 1990 mercury emissions were reduced 70% by 2005, which is equivalent to 76% statewide emissions reduction goal, leaving 24% of the emissions reductions goal remaining. Going from 3,341 lbs mercury emissions in 2005 to the emissions goal of 789 lbs constitutes another 76% reduction in mercury emissions.

TMDL Implementation To achieve the mercury reductions goals, Minnesota will develop a detailed implementation plan. An implementation plan is not required in a TMDL; it is developed after the TMDL plan is approved by the USEPA. A section on projected implementation is included in this report to inform the public and to aid in the discussion on reasonable assurance. The implementation and reasonable assurance sections summarize initiatives that the MPCA believes have already reduced fish contamination in Minnesota and will maintain a path of reduced fish contamination in the future. Although wastewater point sources are very minor contributors to the total mercury load, the MPCA will continue to pursue mercury reductions from these sources through mercury minimization plans and other permit conditions. For mercury emission sources, sector-specific reduction milestones are presented, along with an outline of regulatory and non-regulatory mercury reduction strategies to be considered in the detailed implementation planning. The Great Lakes Initiative (GLI) requires wastewater dischargers in the Lake Superior basin to meet a mercury water quality standard of 1.3 ng/L and implementation of this mercury TMDL does not in any way supercede or conflict with the GLI requirements.

Abbreviations

- number

% - percent

A – area

b – bioavailability factor

C – mercury concentration

Cl – chloride

D_y – annual air deposition of mercury

d – day

DA – Driftless Area ecoregion

DDT – dichlorodiphenyltrichloroethane

ECOS – Environmental Council of States

EPA – US Environmental Protection Agency

EU – European Union

FCA – fish consumption advisory

g – gram

Hg – mercury

kg – kilogram

km – kilometer

L_{air} – air deposition mercury load

L – liter

LA – load allocation

LaMP – Lake area Management Plan

lb – pound

MACT – Maximum Achievable Control Technology

MDH – Minnesota Department of Health

MDNR – Minnesota Department of Natural Resources

Metro – the Twin Cities Metropolitan Area

MFCA – Minnesota Fish Consumption Advisory

mg – milligram

Minn R ch – Minnesota Rules Chapter

Minn R p – Minnesota Rules part

MMP – mercury minimization plan

MOS – margin of safety

MPCA – Minnesota Pollution Control Agency

NCHF – North Central Hardwood Forest ecoregion

NE – northeast

ng – nanogram

NGP – Northern Glaciated Plains ecoregion

NLF – Northern Lakes and Forests ecoregion

NMW – Northern Minnesota wetlands ecoregion

NP_{55} – standard length Northern Pike – 55 cm

NPDES – National Pollutant Discharge Elimination System

NPSL – nonpoint source load

PCBs – polychlorinated biphenyl compounds

POPs – persistent organic pollutants

ppm – parts per million

PSL – point source load

Q – flow

r – runoff coefficient or delivery ratio

RF – reduction factor

RRV – Red River Valley ecoregion

SA – surface area

SRB – sulfate reducing bacteria

Subp – subpart

SW – southwest

T – temperature

TSL – total source load

TMDL – Total Maximum Daily Load

USGS – United States Geological Survey

WCBP – Western Corn Belt Plains ecoregion

WE_{40} – standard length Walleye – 40 cm

WLA – wasteload allocation

WQS – water quality standards

WWTP – wastewater treatment plant

yr – year

Definition of Terms

Air Sector – sources that emit wastes into the air

Anthropogenic Mercury Emissions – the mobilization or release of geologically-bound mercury by human activity that results in a mass transfer of mercury to the atmosphere.

Atmospheric deposition – the mass transfer of gaseous, aerosol, or particulate contaminant from the atmosphere to the earth's surface (see mercury dry deposition and mercury wet deposition)

Bioaccumulation – increase in contaminant concentration through a food web; includes uptake through food and water or air. Bioconcentration refers to uptake only through the water or air, not food. Biomagnification is increase in contaminant concentration between trophic levels.

Chlorophyll-*a* – a pigment in green plants, including algae. The concentration of chlorophyll-a, expressed in weight per unit volume of water, is a measurement of the abundance of algae.

***de minimis* –** insignificant; reference to the phrase *de minimis non curat lex*: "The law does not concern itself with trifles;" - a principle of law, that even if a technical violation of a law appears to exist according to the letter of the law, if the effect is too small to be of consequence, the violation of the law will not be considered as a sufficient cause of action, whether in civil or criminal proceedings. For the mercury TMDL, wastewater point sources are considered *de minimis* if they represent less than one percent of the TMDL of mercury to the region.

Dystrophic – a high concentration of dissolved humic organic matter in the water, causing the water to have a brown color; also, typically low in nutrients.

Ecoregion – an area of relative homogeneity in ecological systems based on land use, soils, land and surface form, and potential natural vegetation.

Fish consumption advisory – guidelines issued by Minnesota Department of Health for how often certain fish can be safely eaten, based on the data collected in Minnesota's fish contaminant monitoring program.

Global Scale – refers to emissions transported on a global scale; it does not refer to the sum of all emissions on Earth, but rather that portion of total emissions that are transported around the globe.

Great Lakes Initiative (GLI) – the Great Lakes Water Quality Initiative required states in the Great Lakes region to adopt strict water quality standards for bioaccumulative chemicals of concern, which includes a mercury water quality standard of 1.3 ng/L. Minn. R. ch. 7052 codified the GLI requirements and is, therefore, referred to in Minnesota as the GLI rule.

Half-life – The time required for the amount of a substance to decrease to half its initial value (see Residence Time).

Hot spot – refers to a mercury concentration in fish or water that is obviously (1) higher than other concentrations in the area and (2) caused by a local source.

Impaired water – a water body that does not meet applicable water quality standards or fully support applicable beneficial uses, due in whole or in part to water pollution from point or nonpoint sources, or any combination thereof.

LaMP – Lake areawide Management Plan – a watershed management plan required for each of the Great Lakes.

Local scale – A relative term, used to describe the area within which emissions can travel in one diurnal cycle (generally within 100 km of a source). Local influences are characterized by measurable concentration gradients with relatively large fluctuations in air concentrations caused by meteorological factors such as wind direction (Expert Panel 1994)

Lognormal – a variable, such as mercury concentration in fish, that has a skewed distribution (median is much lower than the mean), but the logarithms of the values have more of a normal distribution (mean and median are approximately equal).

MACT – Maximum Achievable Control Technology—a pollution regulatory program administered through the USEPA.

Mechanistic model – a computer model that is based on known physical, biological, and chemical processes.

Mercury dry deposition – mass transfer of gaseous, aerosol, or particulate mercury species from the atmosphere to the earth's surface (either aquatic or terrestrial, including trees and other vegetation) in the absence of precipitation.

Mercury wet deposition – mass transfer of dissolved gaseous or particulate mercury species from the atmosphere to the earth's surface (either aquatic or terrestrial) by precipitation.

Methylation (Methylated) – process of adding a methyl (CH_3^-) group to a mercury ion (Hg^{2+}). Methylation can occur either biotically or abiotically, but sulfate-reducing bacteria are considered the primary methylators in aquatic systems (i.e., wetlands and lakes).

Methylmercury – CH_3Hg^+ or MeHg – a cation that is the biologically active form of mercury; it has a very high affinity for sulfur-containing compounds, such as the amino acid cysteine; this is the form of mercury that accumulates in fish and is toxic to humans and wildlife.

National – for the purposes of this TMDL, national is the United States except Minnesota; thus, mercury emission sources include Minnesota sources, national sources, global sources, and natural sources.

Natural mercury emissions – mobilization or release of geologically-bound mercury by natural biotic and abiotic processes that result in mass transfer of mercury to the atmosphere.

Nonpoint sources – diffuse sources of pollution to water from land use or atmospheric deposition of pollutants.

Oligotrophic – low nutrient concentration, resulting in low plant and animal productivity. Many lakes in northern Minnesota are oligotrophic. The contrasting condition is eutrophic – high nutrient concentration and biological productivity.

Point sources – wastewater discharges and all other pollutant sources that enter the receiving water through a pipe or channel.

Reference Dose (RfD) – an estimate of a daily oral exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime.

Regional scale – a relative term, defining that area requiring more than one diurnal cycle emission transport time (about 100 to 2000 km from a source). The regional scale describes areas sufficiently remote or distant from large emission sources so that concentration fields are rather homogeneous, lacking measurable gradients (Expert Panel 1994).

Reserve capacity – pollutant load allocation to account for uncertainty and future growth.

Residence time - average time that a substance resides in a designated area (e.g., atmosphere or water body).

Standard length fish – a set total fish length that is used to compare mercury concentrations among lakes and over time. The standard lengths used by the MPCA are 55 cm northern pike (NP55) and 40 cm walleye (WE40). Mercury concentrations for a standard length fish are determined from a linear regression of measured mercury fish tissue concentration versus fish length.

Steady state – occurs when there is a balance between inflows and outflows of a system; also referred to as dynamic equilibrium; for the mercury TMDL, this refers to a condition when inflows and outflows are not changing rapidly with respect to changes fish tissue concentrations, which can take 5-10 years to respond to changes in mercury load.

Taconite – low-grade iron ore processed by crushing and concentrating to yield a pellet for use in iron smelters. Taconite has low mercury concentrations but large volumes of the material are heated during processing, which releases significant quantities of mercury into the atmosphere.

TMDL – Total Maximum Daily Load. The maximum amount of a pollutant that a waterbody can receive and still meet water quality standards. TMDL also refers to the process of allocating pollutant loadings among point and nonpoint sources.

1 Introduction

Mercury is a toxic pollutant and eating mercury-contaminated fish is the primary route of exposure for most people and wildlife. Mercury has accumulated in fish throughout the world because of human activities that emit mercury to the environment. Even lakes in natural pristine areas contain fish with high mercury concentrations, because mercury is deposited from the atmosphere and can travel long distances from its emission source.

Two thirds of the waters on Minnesota's 2004 Impaired Waters List are impaired because of mercury concentrations in fish or water (Figure 1). Waters are listed as impaired if mercury in fish tissue is greater than 0.2 ppm, which corresponds to a fish consumption advisory threshold for sensitive populations[1] that is more restrictive than one meal per week. Waters are also listed as impaired if mercury in water exceeds the water quality standard for mercury: 1.3 ng/L in the Lake Superior Basin and 6.9 ng/L elsewhere in the state. The 1239 impairments by mercury consist of 820 lake impairments and 419 river impairments. Twelve lakes and 20 river reaches are impaired for mercury in fish tissue and in the water column; 808 lakes and 399 river reaches are impaired because of fish tissue only.

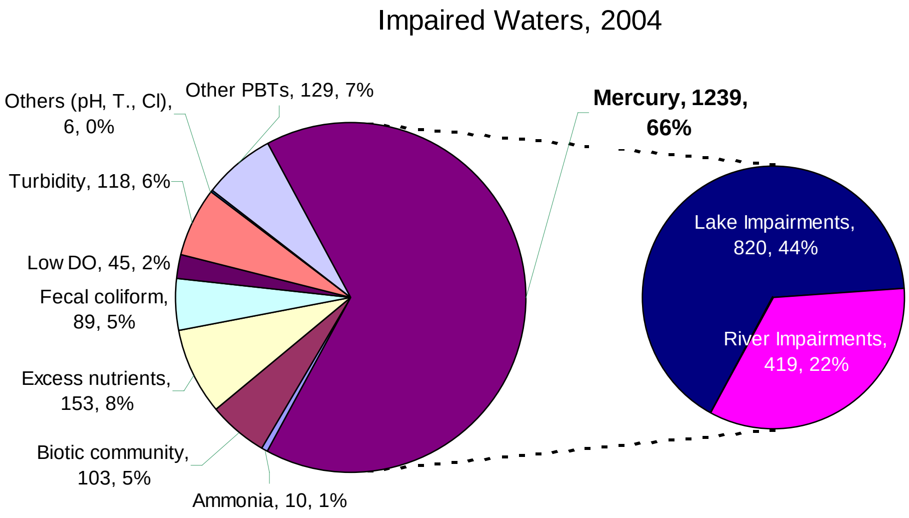

Figure 1 Impaired Waters by Pollutant for 2004 303(d) List

To ensure the continued good health of people that eat fish in Minnesota, the MDH issues guidelines—Minnesota Fish Consumption Advisory (MFCA)—for how often certain fish can be safely eaten (MDH 2004). The MFCA is strictly advisory, the goal being to help people that eat fish make intelligent decisions on which fish to eat and which to avoid. In contrast, the 303(d) [Impaired Waters] List identifies waterbodies that do not meet legally enforceable water quality standards, requiring a study of the total maximum daily load (TMDL) that is necessary to meet the water quality standard.

This report describes the statewide, regional approach that Minnesota has taken to developing total maximum daily loads for mercury. The two regions—Northeast and Southwest—provide clear distinctions for assessing mercury loads to surface waters based on differences between the regions in watershed transport processes and mercury bioavailability.

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[1] Women who are pregnant or intending to become pregnant and children under 15 years old.

2 Background Information

2.1 Mercury Cycle

Mercury has unique properties as a multi-media pollutant, cycling through the air, water, land, and biota. The Mercury Study Report to Congress (USEPA 1997) gave the following summary of the mercury cycle:

Mercury cycles in the environment as a result of natural and human (anthropogenic) activities. The amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age. Most of the mercury in the atmosphere is elemental mercury vapor, which circulates in the atmosphere for up to a year, and hence is widely dispersed and transported thousands of miles from the likely sources of emission. Most of the mercury in water, soil, sediments, or plants and animals is in the form of inorganic mercury salts and organic forms of mercury (e.g., methylmercury). The inorganic form of mercury, when either bound to airborne particles or in a gaseous form, is readily removed from the atmosphere by precipitation and is also dry deposited. … Even after it deposits, mercury commonly is emitted back to the atmosphere either as a gas or associated with particles, to be re-deposited elsewhere. As it cycles between the atmosphere, land, and water, mercury undergoes a series of complex chemical and physical transformations, many of which are not completely understood.

Mercury accumulates most efficiently in the aquatic food web. Predatory organisms at the top of the food web generally have higher mercury concentrations. Nearly all the mercury that accumulates in fish tissue is methylmercury. Inorganic mercury, which is less efficiently absorbed and more readily eliminated from the body than methylmercury, does not tend to bioaccumulate.

A link in the aquatic food chain: a walleye pursuing a yellow perch

2.2 Data Collection and Assessment

This section describes how data are collected and assessed for possible inclusion in the 303(d) list. Essentially all of the discussion is excerpted from the MPCA TMDL guidance document (MPCA 2004a).

The MPCA uses data collected over the most recent 10-year period for all the water quality assessments, except in the case of fish contaminated with mercury. There is no age limit for the use of mercury fish tissue data, for the following reasons. A state-wide trend analysis of mercury fish tissue concentrations measured over the last 10 – 15 years indicates only a very slight average rate of decline of about one percent per year. This is not a large enough downward trend to justify using only the latest 10 years of data. Also, there have been no significant changes in sampling or analytical procedures associated with the fish tissue data that would invalidate the older data. It would not be justifiable to remove a waterbody from the 303(d) list simply because the mercury fish tissue data for that waterbody were collected more than 10 years ago. With the increased interest in evaluating trends in mercury contamination, more lakes and rivers are being resampled. If new data indicates a waterbody is no longer impaired, it is removed from the impaired waters list (i.e., “delisted”). No mercury-impaired water has been delisted.

The fish contaminant monitoring program is a multi-agency program that includes the MPCA, the Department of Natural Resources (MDNR), the Department of Agriculture, and the Minnesota Department of Health (MDH). The primary purpose of the fish contaminant monitoring has been to support MDH’s fish consumption advisory. Fish contaminant data are also used by the MPCA to determine where site-specific studies are needed, to help identify sources of pollutants, and to look for trends in fish tissue levels. Each year some waterbodies are sampled for the first time and some waterbodies are re-sampled. Sample locations are determined by the following:

MPCA Biomonitoring Unit's Mike Feist holding a stream resident

- Where MDNR personnel will be conducting fish population surveys,
- Waterways where fishing pressure is relatively high,
- Where previous data are more than ten years old, or
- Where information is needed for special studies or trend analysis.

Minnesota has been collecting fish for mercury and polychlorinated biphenyls (PCB) tissue analysis since the 1970s. Over the years other bioaccumulative pollutants, such as DDT, dioxins and toxaphene have been analyzed in fish tissue samples, but only at very limited locations where potential problems were suspected.

The edible portion, which is a skin-on fillet, is prepared in the MPCA fish processing lab by DNR personnel. Currently, fish samples are analyzed by the Department of Agriculture analytical lab. Since fish bioaccumulate these pollutants, concentrations below method detection limits are not usually an issue. When they do occur, one half of the method detection limit (less-than value) is used in the assessments. The data for each lake or river reach are separated by species and by individual size classes: 5-15, 15-20, 20-25, 25-30 and 30 + inches. Data collected in the five-year period that includes the most recent sampling is averaged. That is, the assessment program identifies the most recent data point, then searches back five years for additional data from the same waterbody, same species, same size class, and averages them.

Fish can be very mobile and difficult to attribute to a discrete portion of a lake or river reach; therefore, all fish tissue information from a lake are aggregated unless there is evidence to show that fish from certain parts of a lake are isolated and may be exposed to different levels of contamination. For rivers, fish are collected with nets or electrofishing gear in a range of river miles generally not more than five miles apart. Sampled sections of a river are associated with river reaches in the USGS hydrologic unit code system. However, fish tissue data from one or more sampling station may be considered representative of more than just the reach from which they were collected. Adjacent river reaches may be listed as well as the reach from which the fish were collected based on general information about the home range of the species, location of upstream or downstream fish barriers such as falls and dams, and significant river tributaries.

MDNR staff processing fish samples for mercury analysis

2.3 Other State and Federal Regional Mercury TMDLs

Other mercury TMDLs have been completed prior to completion of this TMDL for mercury-impaired waters in Minnesota. This section provides a synopsis of some of those completed mercury TMDLs.

TMDLs for Lakes Listed for Mercury in Fish Tissue for the Ouachita River Basin, Arkansas – There are fish consumption advisories (FCA) throughout the basin (FTN 2003). The Mercury Action Level in Arkansas is 1.0 mg/kg; the target goal is 0.8 mg/kg, using a 20% margin of safety.

The TMDL had a two-step approach. The first step estimated the mercury loads from all sources. The second step, using largemouth bass tissue mercury concentration, estimated the reduction needed to meet water quality standards (WQS). A linear relationship was assumed between mercury in fish and mercury loading. The fish tissue mercury reduction goals were used to determine the load reductions necessary.

Less than 1% of the existing mercury load is from water point sources. Estimated reductions in mercury loading to the basin resulting from the implementation of emission reduction regulations and erosion BMPs were calculated.

Mercury TMDLs for Subsegments Within Mermentau & Vermilion-Teche River Basins, Louisiana – the reaches in these basins are listed because of excess mercury in fish tissue (USEPA 2001a). The regional approach is justified because atmospheric deposition of mercury is the predominant in these basins. They determined a 67% reduction in mercury in fish tissue is required to meet WQS. They assumed a linear relationship between loading reductions and fish tissue reductions. A ten year implementation schedule was projected. The Binational Toxics Strategy sets a national challenge of a 50% reduction of mercury releases by 2006, so that is the interim goal. Because of water body mercury recycling and runoff from land surfaces, after deposition reduction goals are met, it will be several decades before attainment is met.

Mercury TMDLs for Little River and Catahoula Lake Watershed, Louisana – these reaches are not meeting WQS because of excess mercury in fish tissue (Parsons 2003). An endpoint of 0.5 mg/kg methylmercury in edible fish tissue is the target. An estimated 99.5% of current mercury loading is from atmospheric deposition. Loadings must be reduced by 32% to meet WQS. Point sources are less than 1% of the load, so are given no reduction goal. Since conservative assumptions are made throughout, there is no explicit margin of safety. The Binational Toxics Strategy reductions of 50% will easily cover the 32%, according to the TMDL.

Total Maximum Daily Load [TMDL] for Total Mercury in Fish Tissue Residue in the Middle & Lower Savannah River Watershed, Georgia – EPA interpreted Georgia's WQS and has determined that the applicable WQS for total mercury in the ambient water of the Savannah River Basin is 2.8 ng/L (USEPA 2001b). At this concentration, or below, fish tissue residue concentrations of mercury will not exceed 0.4 mg/kg, which is protective of the general population from consumption of freshwater fish. The TMDL was developed by using modeling. Ninety-nine percent of the mercury load is from atmospheric deposition. Based on the modeling, a 44% reduction goal is required to achieve a water column concentration of 2.8 ng/L. Water point sources will be required to create mercury minimization plans.

3 Applicable Water Quality Standards

3.1 Mercury Numeric Water Quality Standards

Mercury numeric water quality standards are based on total (particulate + dissolved) concentrations and thus, total mercury measurements in water are used in assessments. Minnesota has two Class 2 water quality standards for total mercury: the human health-based statewide standard in Minn. R. ch. 7050 and the wildlife-based standard applicable to just the waters of the Lake Superior Basin in Minn. R. ch. 7052. These standards are:

- 6.9 ng/L. chronic standard, Minn. R. pt. 7050.0222
- 1.3 ng/L. chronic standard, Minn. R. pt. 7052.0100
  (ng/L = nanogram per liter, or parts per trillion)

Minnesota R. ch. 7052 is the Great Lakes Water Quality Initiative (GLI), which focuses on the reduction of bioaccumulative toxic chemicals in the Great Lakes ecosystem as a whole.[2] The GLI was mandated by a 1987 amendment to the Clean Water Act; it was promulgated as a federal rule by EPA in 1995 and adopted in Minnesota in 1998. The GLI has been adopted by all eight Great Lakes States, including six states in Region 5. The GLI requires wastewater dischargers in the Lake Superior basin to meet a mercury water quality standard of 1.3 ng/L and implementation of this mercury TMDL does not in any way supercede or conflict with the GLI requirements.

To accurately measure the low levels of mercury in natural waters, the MPCA began using clean sampling techniques (EPA Method 1669) and low-level analysis for mercury (EPA Method 1631 or equivalent) in 1996; only data collected in this manner are used to assess mercury concentration in water. Mercury levels are assessed by comparing concentrations in water to the ambient standards shown above, and by assessing the mercury in fish tissue directly (described below). The analysis of mercury in fish tissue does not require as sensitive a method as used for water, because of bioaccumulation, and the mercury detection limits for fish tissue have not significantly changed since fish tissue analysis began in the mid-1960s.

In 2001, the USEPA issued a fish tissue residue criterion for methylmercury to protect human health (USEPA 2001c). The federal methylmercury fish tissue criterion is 0.3 mg/kg. The fish tissue criterion is expected to be used by states to establish or update water quality standards. The USEPA chose to express the water quality criterion as a fish tissue value rather than a water column value because consumption of fish is the primary route of human exposure. MPCA is proposing to adopt this criterion in addition to the 6.9 ng/L mercury water quality standard; however, Minnesota's mercury standard would be 0.2 mg/kg because of higher fish consumption rates in the state (See Section 4.3.2 and 4.3.3 for more details).

3.2 Fish Consumption Advisory

To ensure the continued good health of people that eat fish in Minnesota, the Minnesota Department of Health (MDH) issues guidelines for how often certain fish can be safely eaten, based on the data collected by the fish contaminant monitoring program. These guidelines are called the Minnesota Fish Consumption Advisory (FCA) (MDH 2004). The MDH, with the help of extensive EPA toxicity and risk assessments for mercury and PCBs, establishes the concentrations of contaminants in fish that trigger the various levels of advice – from "unlimited consumption" to "do not eat" (Table 1). The FCA is strictly advisory, the goal being to help people that eat fish make decisions on which fish to eat and which to avoid. Prior to 2001, the MDH published in booklet form a list of all the lakes and rivers that had fish tested and the subsequent consumption advice for those lakes. As of 2001, the list of individual water bodies is available on the MDH web site, but it is no longer published as a booklet. Instead, the MDH publishes a brochure giving general advice applicable to all fishing lakes (and rivers) in Minnesota, regardless of whether the fish from a given lake or river have been tested; exceptions with more restrictive consumption advice are listed individually.

Table 1 Fish Tissue Mercury Concentrations for MDH's Levels of Consumption Advice

	Mercury Concentration in Fish, ppm			
MFCA for Mercury	< 0.05	0.05 - 0.2	>0.2 - 1.0	> 1.0
Consumption Advice:*	Unlimited	1 meal/week	1 meal/month	Do not eat

*Consumption advice for young children and women of child-bearing age.

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[2] Minn. R. ch. 7052.0210 imposes restrictions on mixing zones for bioaccumulative chemicals of concern (BCCs). Allowance for mixing zones for BCCs are scheduled to end in 2007 (See Section 10.3).

The MDH fish consumption advice and the MPCA impairment determinations are based on the same EPA-derived reference dose. Reference dose, expressed in units of daily dose, is an estimate of the daily exposure to human populations, including sensitive sub-populations, that is likely to be without appreciable risk of deleterious effects over a lifetime. A good summary of the toxicity assessment and derivation of the reference dose for mercury is included in the most recent USEPA mercury criterion document (EPA 2001c).

While the FCA is not mandatory, the 303(d) list of impaired waters is mandatory, listing waterbodies that do not meet legally enforceable water quality standards, and requiring Total Maximum Daily Load studies of those impaired waters.

Contaminants in fish can be a threat to wildlife consumers of fish and other aquatic organisms as well as humans. Minnesota does not have a program to analyze whole fish samples for the purpose of assessing risks to wildlife; however, the water quality standard for the Lake Superior basin (1.3 ng Hg/L) is based on wildlife toxicity of mercury via fish consumption.

3.3 Basis for Assessment of Fish Contaminants – Narrative Standards

3.3.1 Minnesota Rule Chapter 7050

The basis for assessing the contaminants in fish tissue is the narrative water quality standards and assessment factors in Minn. R. pt. 7050.0150, subp. 7 which is quoted below:

Subp. 7. ***Impairment of waters relating to fish for human consumption.*** *In evaluating whether the narrative standards in subpart 3, which prevent harmful pesticide or other residues in aquatic flora or fauna, are being met, the commissioner will use the residue levels in fish muscle tissue established by the Minnesota Department of Health to identify surface waters supporting fish for which the Minnesota Department of Health recommends a reduced frequency of fish consumption for the protection of public health. A water body will be considered impaired when the recommended consumption frequency is less than one meal per week, such as one meal per month, for any member of the population. That is, a water body will not be considered impaired if the recommended consumption frequency is one meal per week, or any less restrictive recommendation such as two meals per week, for all members of the population. The impaired condition must be supported with measured data on the contaminant levels in the indigenous fish.*

3.3.2 Selection of Single Fish Meal-Per-Week Impairment Threshold

The EPA has promulgated a methylmercury criterion for fish tissue because the consumption of fish is the most important route of exposure of mercury to humans. The two aspects of a criterion are toxicity and exposure. The MPCA relies on the assessments of mercury toxicity to humans by the experts within EPA and MDH. Exposure varies with how often people eat fish and with the mercury concentrations in the fish they eat. The EPA assumes people eat 17.5 grams per day for purposes of calculating their human health-based aquatic life criteria (EPA 2000c). This generic assumption applies to everybody in the U.S. The MPCA uses a higher consumption rate for determining exposure because of the prevalence and importance of sport fishing in Minnesota. Minnesota human health-based water quality standards are calculated assuming people eat 30 grams of fish per day, which is the 80th percentile fish consumption rate of sport-caught fish for the angling population, based on several surveys of the fish eating habits of upper Midwest anglers (not the population as a whole) (MPCA 2000).

The surveys from which the 30 g/d value was derived indicate that less than 20 percent of anglers and less than 95 percent of the whole population in the upper Midwest eat more sport-caught fish than one meal-per-week averaged over a lifetime (MPCA 2000). A more recent survey of fish consumption habits of people living in Minnesota and North Dakota suggests that 30 g/d may be more protective than these earlier surveys indicate (EERC 2001). The 95th percentile consumption rates of sport-caught fish for all Minnesotans with fishing licenses reported in the EERC survey is 30.4 g/d (32.1 g/d in a lognormal distribution).

Thirty grams per day equals about a half-pound meal per week (0.463 pounds/week). The single fish meal-per-week consumption rate (or 30 g/d) is the basis for all Minnesota human health-based water quality standards in both Minn. R. chs. 7050 and 7052. Therefore, "fish consumption" use is supported if it is safe to eat one fish meal per week (over a life time), consistent with the assumption inherent in the numeric water quality standards. In other words, MDH's advice to limit consumption to "no more than one meal-per-week" (or advise that allows more consumption) is not considered an exceedance of the mercury water quality standards, and waterbodies with such advice are not listed as impaired. Advice to limit consumption to less than one meal per week, such as one meal per month, for any member of the population, indicates impairment.

3.3.3 Fish Tissue Criterion for Mercury

Relevant to the assessment of mercury in fish is the issuance by EPA of a revised human health-based water quality criterion for methylmercury (EPA 2001c). This new criterion is unique among all EPA (Clean Water Act section 304(a)) criteria in that the medium for the acceptable mercury concentration is fish tissue rather than water. A fish tissue criterion for mercury is logical because it is fish that are the main source of methylmercury exposure to both humans and wildlife. Also, a tissue-based criterion eliminates the need for a bioaccumulation factor in the criterion calculation, which can be a significant source of uncertainty. The bioaccumulation factor is the ratio of fish concentration to water concentration. The new EPA criterion is 0.3 mg/kg (ppm) methylmercury in fish muscle tissue. Since nearly 100 percent of the mercury in fish muscle is methylmercury, it is essentially a total mercury criterion.

In the determination of the 0.3 ppm criterion, EPA assumes people eat 17.5 grams of fish per day, as mentioned above. If the EPA criterion is re-calculated assuming people eat 30 g/day, the criterion becomes 0.17 ppm. This EPA criterion and the MFCA are both based on the same EPA-derived reference dose of 0.1 µg/kg/day. The difference between the MDH value of 0.2 ppm and the re-calculated EPA criterion of 0.17 ppm, both of which assume a single half pound meal of fish per week, has to do with how the consumption of marine fish is taken into account. The MFCA is advice about eating fish from any source, sport-caught, store-bought, marine or freshwater. The EPA aquatic life criteria (applicable in Minnesota) apply only to freshwater habitats. But, in the calculation of freshwater criteria, EPA assumes people eat a certain amount of marine fish in addition to the 17.5 g/d of freshwater fish. As a result, the freshwater criterion is lowered to allow for this "outside" source of mercury (this is standard procedure in EPA criteria and MPCA standard calculations). Thus, the re-calculated mercury criterion ends up at 0.17 rather than 0.2 ppm. The MPCA believes that the use of 0.2 ppm, rather than 0.17 ppm as the basis for impairment decisions is appropriate for the following reasons:

- EPA rounded the reference dose of 0.1 µg/kg/day to one significant figure; thus, 0.17 and 0.2 ppm could be considered essentially the same number,
- The use by MPCA of the more protective fish consumption amount (30 g/d),
- The use of safety factors in the criterion calculation (again, standard procedure),
- Uncertainties inherent in criteria development, and
- The importance of maintaining consistency in the MPCA/MDH approaches.

4 Minnesota's Regional Approach

4.1 Land Cover and Water Quality

The primary source of mercury to the state's water bodies is atmospheric deposition, which is approximately uniform across the state (See Section 5.1). Mercury concentrations in fish, however, vary widely on both large and small scales. A major factor causing this spatial variability in fish mercury levels is land cover and use, which affects watershed transport of mercury, background water chemistry, and nutrients. The water quality differences, influenced by the nutrient loading, in turn influence the bioavailability of mercury. Wetlands – an important land cover in Minnesota – are important sites of mercury methylation, because sulfate-reducing bacteria (SRB) are the primary methylators and they thrive in wetlands. Additional sulfate to a wetland can stimulate SRB activity, leading to increased mercury methylation (See Section 5.4). Methylmercury associated with dissolved organic carbon released from wetlands is conveyed to surface waters (Driscoll *et al*. 1995). Consequently, wetland density is correlated with mercury concentrations in water and fish (e.g. Greenfield *et al.* 2001, Knights *et al*. 2005). Compared to wetland and forested lands, cultivated lands are typically sources of high suspended solids because of soil erosion. Research in Minnesota has shown mercury is associated with the high suspended solids loads (Balogh *et al* 1997, Balogh *et al*. 1998), although most of the mercury associated with the solids has low bioavailability, because only a small fraction is methylmercury.

A concise literature review of the relationship between lake trophic status and fish mercury concentration was provided by Fink *et al.* (2001). Studying Swedish lakes, Johnels *et al*. (1967) observed that high fish mercury levels were less likely in nutrient-enriched or eutrophic lakes than in unenriched or oligotrophic lakes. The same observation was made by D'Itri *et al.* (1971) and Hakanson (1974); and was attributed to the buffering or dilution effect caused by an increase in suspended solids of biological origin. Several processes may contribute to this "biodilution" effect: (1) increased biomass for sorbing the mercury, (2) increased settling rate of biomass, which removes sorbed mercury from the water column, and (3) increased growth rate of fish, resulting in larger size for a given age class (Norstrom *et al.,* 1976). The biodilution effect has been demonstrated in laboratory microcosms (Pickhardt *et al.* 2002) and in empirical field studies (Chen and Folt 2005)

4.2 Ecoregions

Regional patterns in lake water quality have been documented for ecoregions in Minnesota (Heiskary, Wilson, and Larsen 1987). The State is divided into seven Level III ecoregions (Omernik and Gallant 1988): Northern Lakes and Forest, Northern Minnesota Wetlands, Red River Valley, North Central Hardwood Forests, Northern Glaciated Plains, Western Corn Belt Plains, and Driftless Area. The principle that land use influences water quality has been put to good use in the development of nutrient criteria for the state. Phosphorus criteria have been proposed based on ecoregions (Heiskary and Walker 1988). A similar approach is proposed for this TMDL to assess mercury loading.

For the purposes of the Minnesota mercury TMDL, the Northern Lakes and Forest ecoregion and the Northern Minnesota Wetlands ecoregion are combined to form the Northeast (NE) region; the other ecoregions are combined to form the Southwest (SW) region (Table 2; Figure 2). Differences between the NE and SW regions are described below. Impaired reaches along the NE/SW boundary were examined and some river reaches were reassigned to maintain a TMDL connection with downstream reaches. A Twin Cities Metro region was considered, based on the dominance of urban land use (i.e., impervious area); however, the fish tissue mercury concentrations for the Metro region were the same as the SW; therefore, the Metro region was combined with the SW region (See Section 4.4).

NE and SW regions are distinct in terms of land cover/use and in water quality (Table 2). The NE region is dominated by wetlands and has less than ten percent cultivated land; the SW region, overall, has less than ten percent wetlands and is dominated by cultivated land. The NE region has an abundance of dystrophic and oligotrophic lakes; the latter is evident by the very low total phosphorus and chlorophyll *a* concentrations (Table 2). In contrast, the SW region has chlorophyll concentrations that are 2 to 10 times higher than the NE region levels. The regional differences in mercury transport and transformation processes result in significantly different average fish tissue mercury concentrations (see Section 4.4).

Table 2 Regional Differences in Land Cover and Water Quality

Region	Eco-region	Wetlands (%)	Cultivated (%)	Hay/ Pasture (%)	Lake TP (µg/L)	Stream TP (µg/L)	Lake Chl *a* (µg/L)	Stream Chl *a* (µg/L)
NE	NMW	58.9	8.3	4.6	14 – 27	40 – 90	2-10	3.2
	NLF	26.9	3.6	6.5	14 – 27	20 – 50	2-10	2.1
SW	NCHF	14.9	35.0	23.4	23 – 50	60 – 150	5 – 22	15.8
	RRV	7.3	78.8	6.6	23 – 50	110 – 300	5 – 22	22.1
	NGP	4.6	73.5	16.5	130 – 250	90 – 250	30 – 55	27.1
	DA	4.5	23.8	27.1	N/A	60 – 150	N/A	N/A
	WCBP	2.9	76.8	12.2	65 – 150	160 – 330	30 – 80	23.6

NMW: Northern Minnesota Wetlands
NLF: Northern Lakes and Forests
NCHF: North Central Hardwoods Forests
WCBP: Western Corn Belt Plains
RRV: Red River Valley
NGP: Northern Glaciated Plains
DA: Driftless Area

Land cover data from 1992 (USGS, 1999. Minnesota Land Cover Data Set)
Total phosphorus (TP) and chlorophyll *a* (Chl a) ranges are typical summer lake water quality conditions and typical annual stream water quality conditions for minimally impacted waters (MPCA. 2003. *Comparison of typical Minnesota water quality conditions.* http://www.pca.state.mn.us/publications/wq-s1-02.pdf). Lake data were collected 1985-1988 (Heiskary and Wilson 1989) and stream data were collected 1990-1992 (McCollor and Heiskary 1993).

4.3 Baseline Year: 1990

The baseline year for the Minnesota mercury TMDL is 1990, because (1) the most recent measurement of total (wet and dry) mercury deposition in Minnesota is based on lake sediment cores collected around 1990, (2) it is a well-established baseline for mercury emissions inventories and, most importantly (3) it represents the end of a period when mercury emissions and fish concentrations were most likely in a steady state. Minnesota's Mercury Emissions Inventory (MPCA 2005; see Figure 13 on page 28) and goals established in Minnesota's Mercury Reduction Law are based on the 1990 baseline. The USEPA has also used 1990 as the baseline for presenting U.S. mercury emissions (see Figure 12 on page 27). The Great Lakes Binational Toxics Strategy (http://www.epa.gov/glnpo/p2/bns.html) and the Lake Superior Lakewide Management Plan (http//www.epa.gov/glnpo/lakesuperior/chapter3.html) use 1990 for assessing reductions in mercury releases.

This regional mercury TMDL relies on mercury deposition being in a dynamic equilibrium, or steady state, with mercury in fish. No one knows how long it takes for fish mercury levels to reflect change in mercury use and emissions, but the lag time is most likely more than 5 years, because of lags in mercury disposal, deposition, methylation, and fish growth. There were rapid changes in mercury use and emissions in the 1990s, as mercury use was eliminated in products (paint, golf course fungicides, alkaline batteries) and incinerator emissions were progressively controlled (see Section 5). As a result, one would not expect fish to be in steady state with contemporaneous mercury emissions after 1990. Therefore, this TMDL establishes the relationship between mercury emissions and fish contamination using data from 1990 or earlier, when conditions had been relatively stable for 10 years (see Figure 11 on page 27).

The best estimate of total mercury deposition is based on sediment cores from Minnesota lakes (Swain *et al.* 1992). Total mercury deposition includes wet and dry deposition. Wet deposition is measured through the Mercury Deposition Network (beginning in 1996) at five locations within or very close to Minnesota, as well as other places throughout the country (http://nadp.sws.uiuc.edu/mdn/). Dry deposition cannot be measured directly, like wet deposition. Therefore, it is not possible to measure total mercury deposition directly. The best measure of total deposition generally accepted among scientists is based on sediment core analysis. Based on sediment cores collected in Minnesota lakes, the best estimate for total mercury deposition around 1990 is 12.5 g km^{-2} yr^{-1}. This total mercury deposition value cannot be correlated with wet deposition measurements, because wet

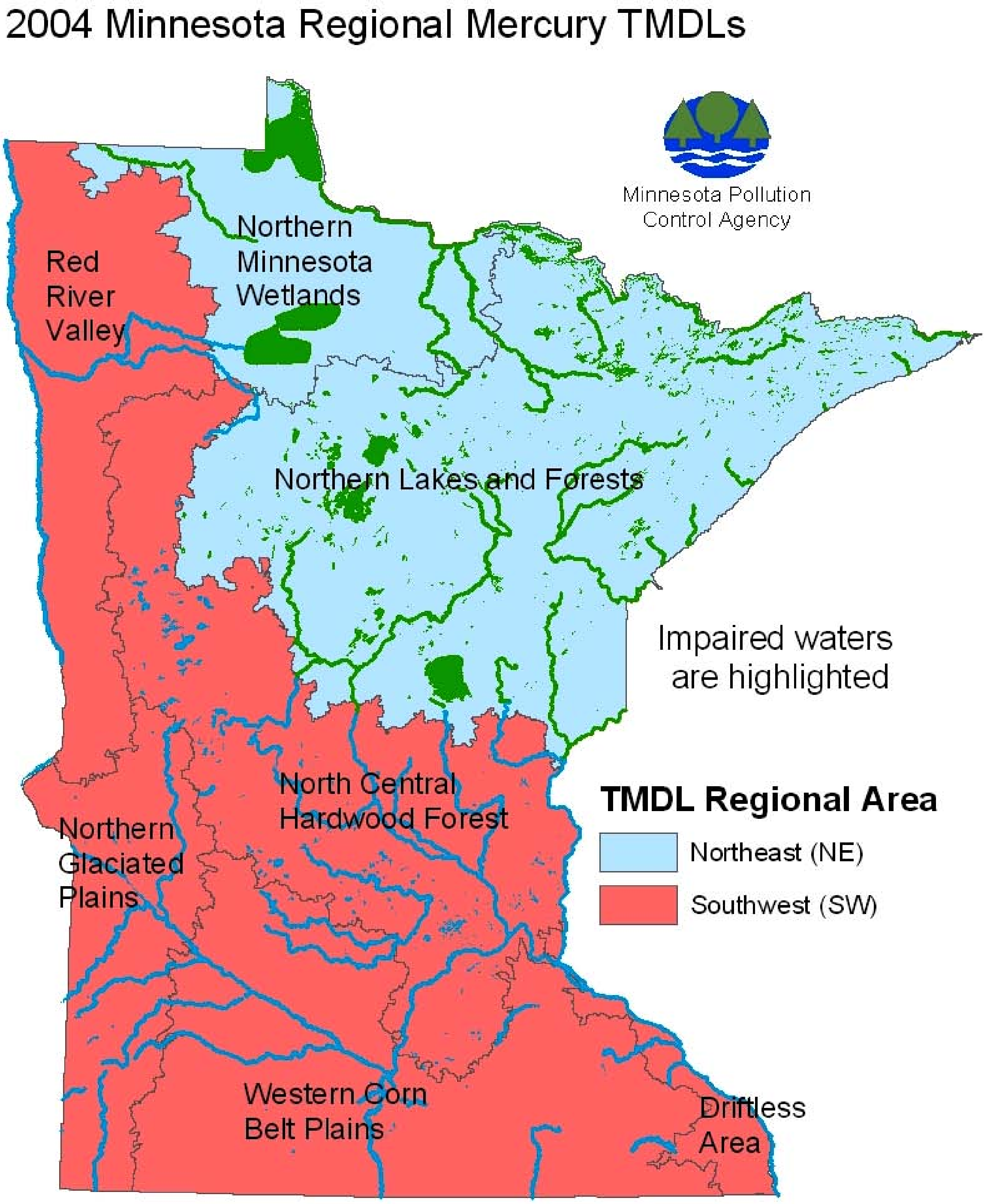


Figure 2 TMDL Regional Areas and Mercury-Impaired Waters

deposition monitoring began later. Therefore, we cannot use the measured wet deposition to derive a reasonable estimate of total mercury deposition for 2000 or 2005. Sediment cores have a time resolution of about ten years. Because of the difficulty measuring total mercury deposition, mercury emissions goals are more practical for the TMDL.

4.4 Target Levels of Fish Mercury Concentration

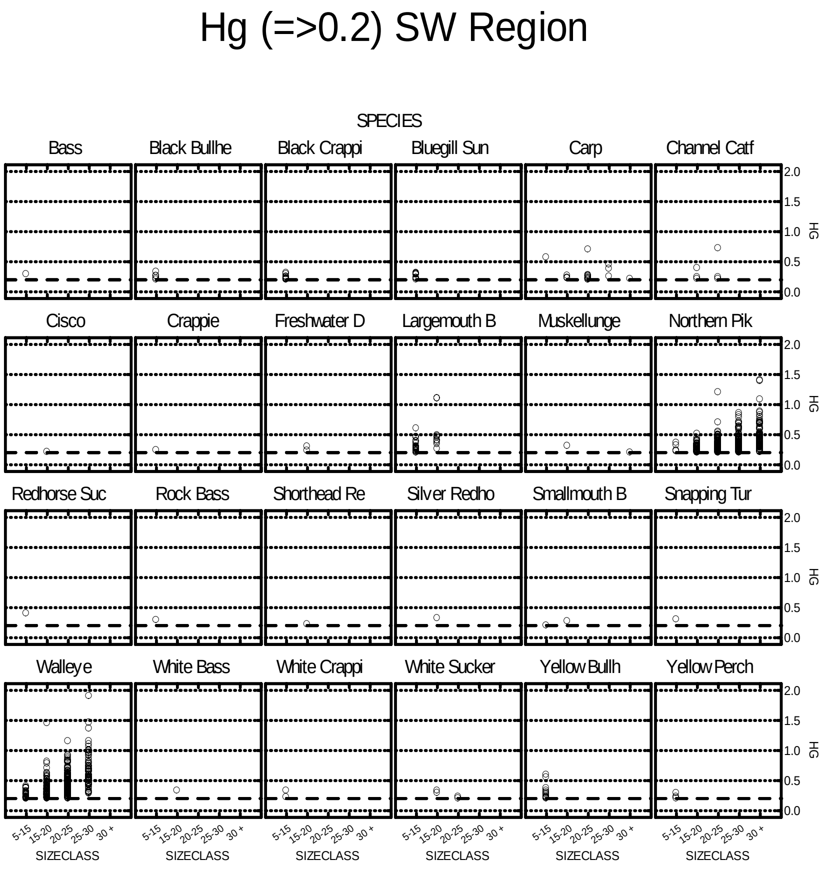
4.4.1 Standard Size Predator Fish

The fish tissue mercury concentration of 0.2 ppm is the mercury TMDL target level. This section describes how the target level is used to derive a mercury reduction factor for each region.

The Minnesota Fish Consumption Advisory (FCA) divides meal advise into five size classes (5-15, 15-20, 20-25, 25-30, 30+ inches); the 0.2 ppm fish tissue mercury concentration is compared to the average for a particular size class of a specific species within a specific water body. For example, a river reach would be considered impaired if the average mercury concentration for the 20-25 inch size class carp was above 0.2 ppm. In the SW region, there are 24 species that had a size class exceed the 0.2 ppm threshold and 32 species in the NE region (Figure 3). Most of these species are only representative of one or two waterbodies. It is not practical to assign a target value for every species for the purposes of this TMDL. Rather, our choice is to use top predators – northern pike and walleye– as the target species, because they have the highest mercury concentrations and are the most common species representing impaired waters. If the top predators meet the water quality standard, then other species and the water column will too.

For temporal and spatial comparisons of mercury concentrations in the top predator fish, we calculate a mercury concentration for a standard length fish. When fish are captured for mercury analysis, the number and size of fish will vary among lakes and among trips to the same lake. Because mercury concentration increases with fish age and fish increase in length with age, comparing the average mercury concentrations among lakes would not account for this relationship to the size of the fish. A sample set from one year could have a higher average mercury concentration than another year because the average fish size was larger the first year; therefore, average concentrations would represent a change in average fish size rather than a real change in mercury concentration.

To account for this size-dependency of mercury concentrations, the mercury concentration is compared for the same size fish, which we refer to as the standard size or standard length fish. One cannot simply select the standard length fish from the collection of fish each time because lengths of collected fish vary from year to year (and lake to lake). The mercury concentration in the standard length fish is calculated for each collection using a linear regression statistical procedure, which gives a mercury concentration per length of fish. The linear regression is done mathematically. It is illustrated by plotting mercury concentration (on the vertical axis) versus the fish length (on the horizontal axis) and drawing a best fit line through the points. The line represents the best estimate of mercury concentration per fish length and can be used to predict mercury concentration for any fish length under the line. In other words, this process allows one to predict the mercury concentration for a pre-determined size of fish based on the available fish data. A collection may have as few as three fish or up to fifty fish, which are used to predict the standard length fish mercury concentration. That single value for the standard length fish can then represent the mercury concentration for the fish from that collection. If the standard length fish mercury concentration decreases between sample collections (and the relationship between age and length of fish does not change), we know that mercury concentrations in that fish species from that water body has decreased and has not changed simply because the collected fish are smaller.



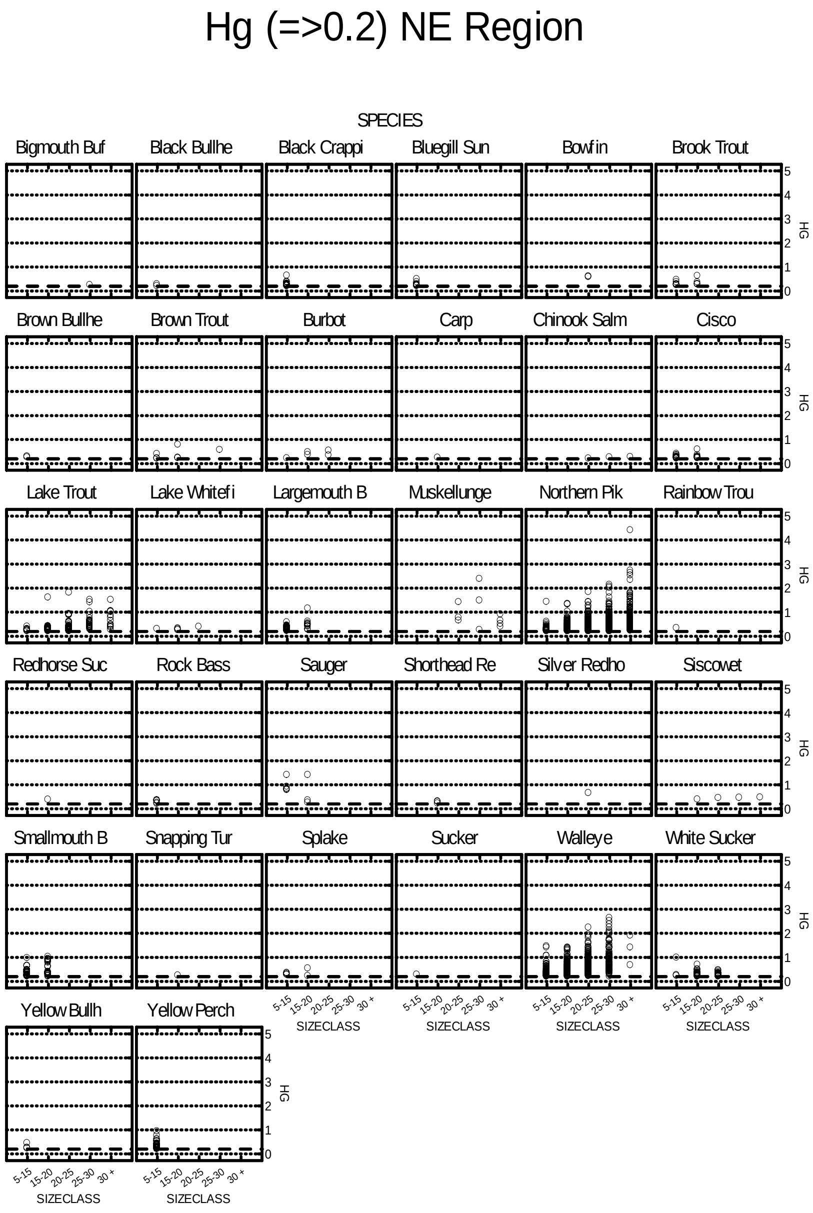


Figure 3 Average Fish Tissue Mercury Concentration Greater Than 0.2 ppm by Species and Size Class

An important aspect of the standard size fish mercury concentration is that it is based on a set of data from a water body, not a single data point; therefore, unlike a single data point, a measure of uncertainty (such as a confidence interval) can be assigned to the value. The measure of uncertainty will vary with the number of data points available and the variability of the data. For future assessments, the standard size fish mercury concentrations can be evaluated to see if there has been a statistically significant change in fish tissue mercury concentrations.

The 55 cm northern pike (NP_{55}) has represented the standard length fish in many scientific mercury studies. Sweden has used the 1-kilogram northern pike—which is approximately equivalent to a length of 55 cm—for mercury studies since the 1960s (Johnels et al. 1967). That standard fish length was used by Sorensen *et al*. (1990) in their study of mercury in northern Minnesota lakes. They compared a range of walleye and northern pike fish lengths and found that a 39 cm walleye had the best correlation to a 55 cm northern pike. The MPCA has continued to use the NP_{55} and a 40 cm walleye (WE_{40}) as the standard length fish to compare among lakes and rivers.

NP_{55} is 21.65 in (~22 in) and WE_{40} is 15.75 in (~16 in); therefore, they are in the 20-25 inch and 15-20 inch size classes used by the FCA. Size class distributions of northern pike and walleye within the two regions indicate the mode (most common or highest frequency) size class for northern pike (NP) in both regions is 20-25 inches and the mode size class for walleye (WE) in both regions is 15-20 inches (Figure 4**Error! Reference source not found.**). Therefore, the standard lengths provide a good correspondence to the most common size class for northern pike and walleye throughout Minnesota.

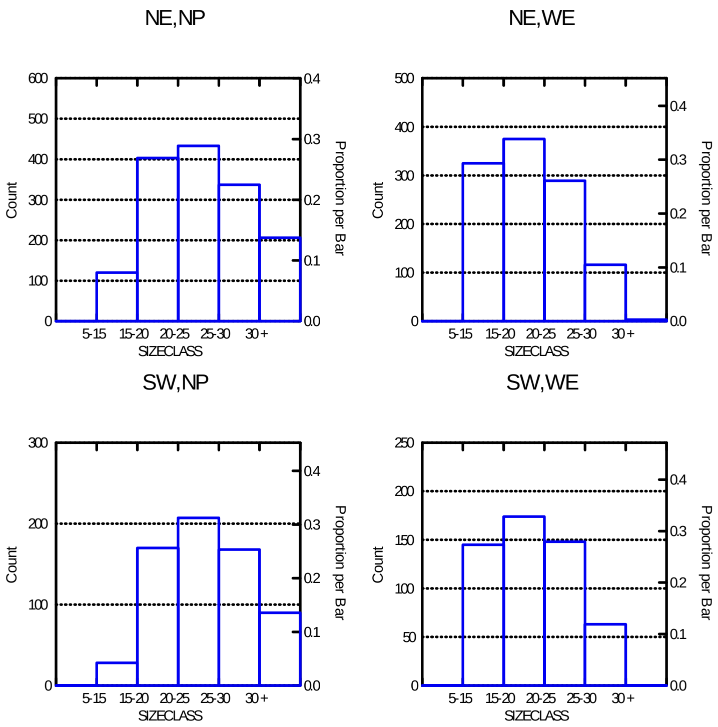


Figure 4 Size Class Distributions by Region (NE, SW) and Species (NP, WE)

4.4.2 Applying the Target Level to the 90th Percentile Fish Concentration

To achieve water quality standards and protect water bodies from impairment, an appropriate statistic must be selected to meet the target level of 0.2 ppm. Rather than using a measure of central tendency, such as the mean or median, the 90th percentile (90%) was selected to provide greater protection. The 90th percentile of samples from a given waterbody has been used as assessment guidance by the USEPA (i.e., no more than 10% of the samples can exceed the standard). Achieving the target level for the 90th percentile of standard size predator fish ensures that 100% of the smaller predator fish and fish at lower trophic levels will meet the target level.

Selecting a higher percentile is not necessary or reasonable. As fish mercury levels are reduced and the 90th percentile approaches 0.2 ppm, the concentration difference between the 90th and the 99th is likely to be very small. Futhermore, the 90th percentile allows for outliers that may have unique circumstances. The outliers can be addressed individually as part of the adaptive watershed management approach to TMDL implementation.

The median (50%) and 90th percentile (90%) mercury concentrations for NP and WE in the two regions are shown in Table 3, based on the period of record (1970-2002). Median mercury concentrations in the 20-25 in size class of NP are similar to the median for the standard length NP for both regions; and the similarity also holds for the 90th percentiles. For WE, the match between mode size class (15-20 in) and standard fish length is not as good as in NP. The WE mercury concentrations are lower, which is not surprising since the standard length WE (16 in.) is near the small end of the 15-20 inch size class and mercury concentrations generally increase with fish length; nonetheless, there is less than a 20% difference between the median and 90th percentile statistics.

Table 3 Mercury Concentrations for Northern Pike (NP) and Walleye (WE) Collected from 1970 to 2002

	Species:	NP		WE	
Region	Size Class (in):	20-25	Std 22 (NP55)	15-20	Std16 (WE40)
NE	N	433	712	375	584
	Median	0.310	0.320	0.318	0.268
	90%	0.645	0.641	0.677	0.631
SW	N	207	265	174	220
	Median	0.190	0.187	0.212	0.185
	90%	0.425	0.368	0.470	0.390

To establish the target level for the baseline year, data from 1988 through 1992 were used to provide a five-year dataset centered around 1990. Five years of fish tissue data provides some accounting for year to year variability in weather, sampling locations, and natural variability in fish populations. The spatial distribution of the fish tissue data reflects distribution of lakes throughout the state; most of the lakes are in the NLF and CHF ecoregions (Figure 5). Many of the lakes had both walleye and northern pike collected and were usually closely matched in fish mercury concentration. The highest mercury concentrations are clearly shown in the northeastern area of the NE region, which includes the Voyageurs National Park and Boundary Waters Wilderness Canoe Area.

Not all mercury impaired waters are included in this mercury TMDL. To ensure that water quality standards are met when the TMDL reduction goal is achieved, only lakes and streams with fish size class mercury concentrations less than 0.572 mg/kg are included (see Appendix A). In other words, only impaired waters with fish size class mercury concentrations that require a reduction of 65% or less are included in this mercury TMDL. MPCA will continue to monitor and assess the other mercury impaired waters, and where feasible, develop waterbody-specific mercury TMDLs.

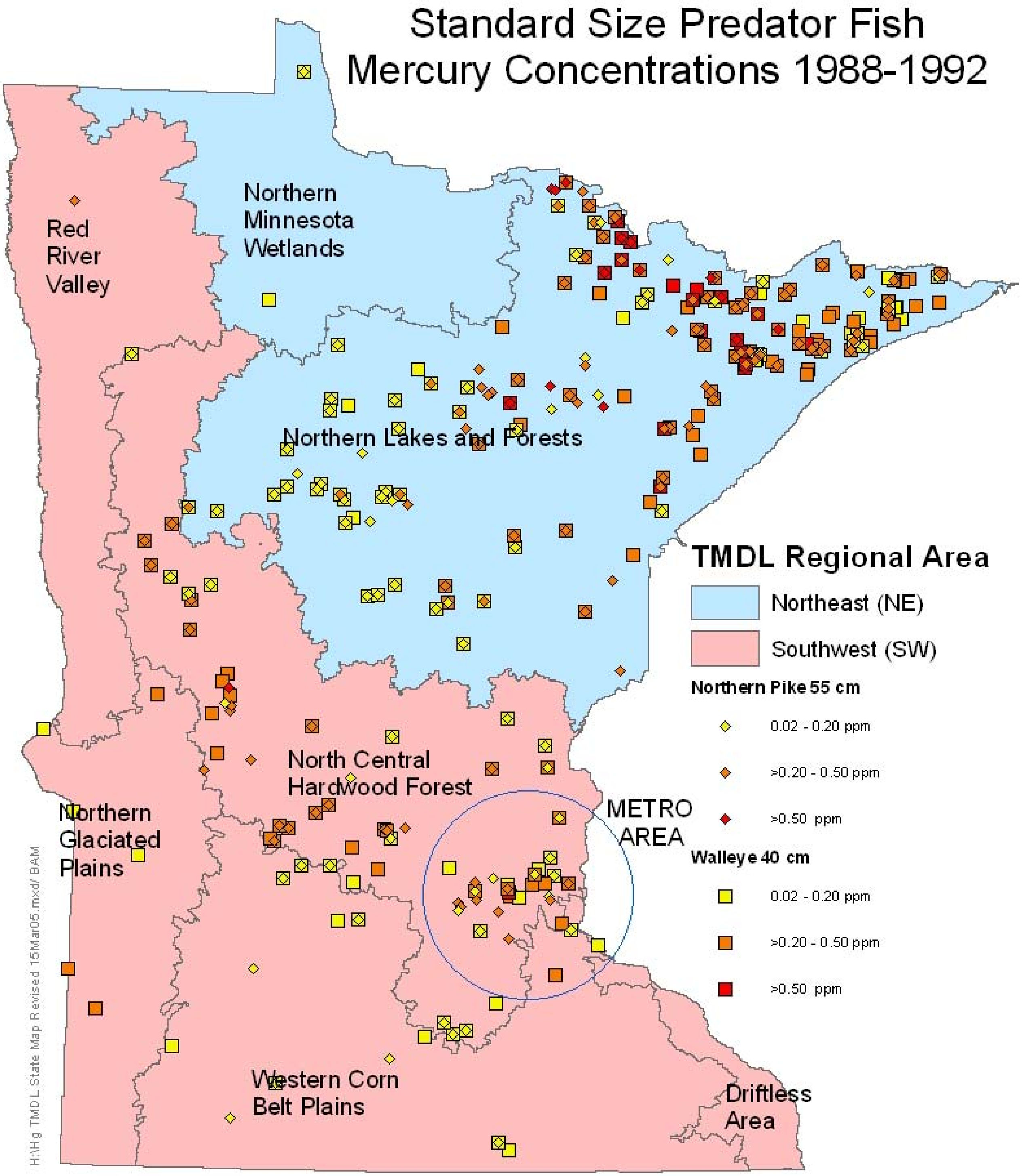


Figure 5 Standard size northern pike and walleye distribution

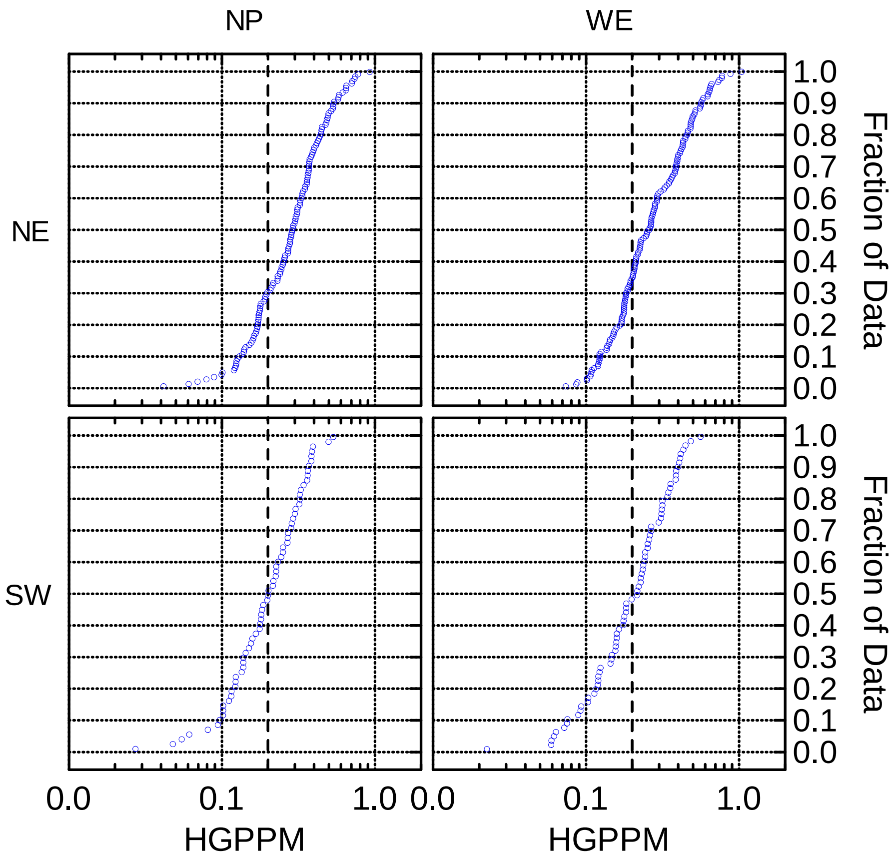


Figure 6 Mercury Distributions by Species and Region for 1988-1992

Figure 6 shows the distribution of mercury concentrations for each standard length species, within each region; the vertical dashed line indicates the 0.2 ppm Hg concentration. Our general goal is to shift the distribution to the left, but for the purpose of assigning a goal or target level, a single statistic is necessary.

The medians and 90th percentiles for 1988-1992 are similar to the whole period of record—1970-2002 (Table 4). The median mercury concentrations in the SW region are actually close to the 0.2 ppm target concentration. Applying the target concentration to the median would clearly not be sufficient to achieve water quality standards in most of the impaired waters within that region.

Table 4 Mercury Concentrations Standard Size Northern Pike and Walleye for 1988-1992

Region	Statistic	NP	WE
NE	N	138	156
	Median	0.293	0.262
	90%	**0.545**	**0.572**
SW	N	66	74
	Median	0.203	0.218
	90%	**0.373**	**0.405**

The 90th percentile mercury concentration provides greater protection than the median and, therefore, was selected as the basis for establishing reduction factors (Table 5). The reduction factor (RF) is the percent reduction needed to achieve 0.2 ppm Hg for the 90th percentile of the standard length fish mercury concentration. Mercury concentrations in standard length walleye ($WE40_{90}$) were higher than standard length northern pike ($NP55_{90}$) in both regions; therefore, the walleye require a greater reduction factor than the northern pike, and achieving the target level for walleye provides a greater level of protection. **Consequently, we selected the more protective reduction factors of 51% for the SW region and 65% for the NE region**.

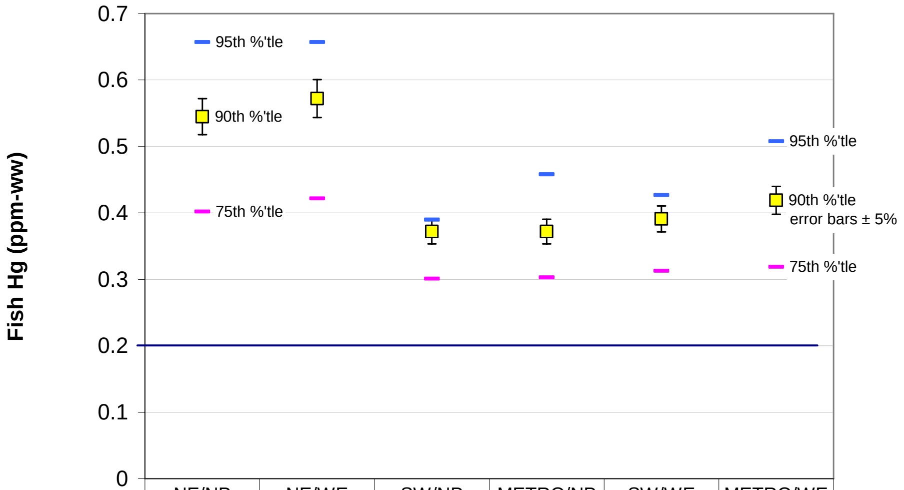
Table 5 Reduction Factors Based on the Baseline Year: 1990

Region	Target Species	Hg* (mg/kg)	RF**
NE	$NP55_{90}$	0.545	63%
	$WE40_{90}$	0.572	65%
SW	$NP55_{90}$	0.373	46%
	$WE40_{90}$	0.405	51%

* 90^{th} percentile for fish mercury data from 1988-1992

** RF: Reduction Factor = $(NP55_{90} - 0.2)/NP55_{90}$ or
= $(WE40_{90} - 0.2)/WE40_{90}$

As mentioned in Section 3, a third, Metro region, defined by the seven county Twin Cities metro area, was considered. The Metro region was separated from the SW region because presumed greater predominance of impervious land cover in the metro region, and potentially higher mercury loading via stormwater. As shown in Figure 7, the SW and Metro had identical 90^{th} percentiles for standard size northern pike (NP), and the walleye (WE) differed by only 0.028 mg/kg (relative percent difference: 7%). The difference between Reduction Factors for SW and Metro WE was only three percent. Given that the Metro region would otherwise be included in SW region and the similarity of the 90^{th} percentiles would result in the same reduction factors, the data from the Metro region were combined with the SW for the final mercury TMDL calculations.



	NE/NP	NE/WE	SW/NP	METRO/NP	SW/WE	METRO/WE
− 95th %'tle	0.657	0.657	0.390	0.458	0.427	0.508
☐ 90th %'tle	0.545	0.572	0.372	0.372	0.391	0.419
− 75th %'tle	0.402	0.422	0.301	0.303	0.313	0.319

Figure 7 Upper percentiles for northern pike (NP and walleye (WE) in the NE, SW, and Metro Regions

4.4.3 Achieving Water Quality Criteria for Mercury

Water column impairments for total mercury are expected to meet water quality standards when the fish tissue target is reached. Minnesota's water column chronic standards for total mercury are 6.9 ng/L statewide and 1.3 ng/L in the Lake Superior basin. The fish tissue target concentration is 0.2 mg/kg. The 0.2 mg/kg fish tissue target can be divided by a representative bioaccumulation factor (BAF) to get a comparable water column mercury concentration. Our best set of BAFs for fish mercury concentrations and total mercury concentrations comes from 14 lakes representing three geographic areas of Minnesota (Table 6). The lakes representing the agricultural area ("Ag") and the Twin Cities metropolitan area ("Metro") are in the SW region; lakes representing the forested area ("Forest") are in the NE region.

Water column concentrations are based on five samples collected in 2000-2001, and standard size northern pike and walleye mercury concentrations are from 1999-2001. Table 1 shows the geometric mean for the 14 BAFs is 388,424 L/kg.

Using the geometric mean BAF, the calculated water concentration for a 0.2 mg/kg fish mercury concentration is 0.52 ng/L; well below the statewide and Lake Superior basin water quality standards. The fifth percentile of the 14 lake BAFs gives a comparable water column mercury concentration of 1.3 ng/L. Therefore, these results provide reasonable assurance that the water column mercury concentration will be met when the fish tissue target is reached.

Table 6 Bioaccumulation factors for 14 representative lakes in Minnesota

REGION	DOWID	WATERWAY	SP-YR	N	NP_{55} (mg/kg)	Water (median, ng/L)	BAF (NP_{55}/Water)
Ag	07-0053	Duck	NP-00	7	0.034	1.057	32,587
Ag	47-0082	Dunns	NP-99	5	0.256	1.180	216,875
Forest	38-0231	Tettegouche	NP-00	24	0.258	1.090	236,676
Ag	41-0089	Shaokatan	WE-00	22	0.203	0.753	269,495
Metro	62-0056	Owasso	NP-01	24	0.148	0.423	351,046
Forest	38-0242	Wolf	WE-99	14	0.184	0.523	352,242
Forest	38-0068	Windy	NP-99	10	0.708	1.753	404,178
Metro	27-0016	Harriet	WE-00	19	0.231	0.488	473,875
Metro	27-0031	Calhoun	WE-00	24	0.262	0.495	528,474
Metro	27-0039	Cedar	NP-00	6	0.356	0.599	594,455
Ag	47-0088	Richardson	NP-99	15	0.305	0.496	614,931
Forest	29-0015	Williams	NP-99	24	0.264	0.425	622,780
Metro	27-0137	Christmas	NP-01	26	0.223	0.229	971,214
Forest	29-0043	Shingobee	NP-00	7	0.342	0.240	1,426,490
				Min:	0.034	0.229	**32,587**
				Max:	0.708	1.753	**1,426,490**
				Geomean:	0.270	0.696	**388,424**

5 Source Assessment and Trends

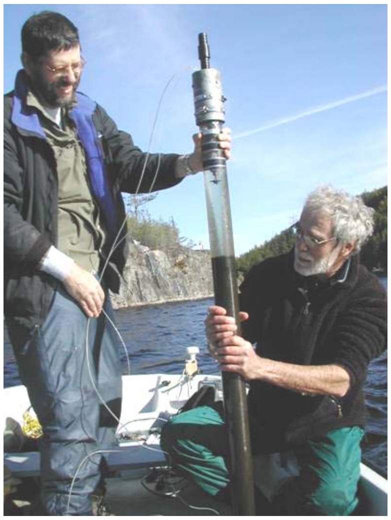
5.1 Sources of Mercury in Minnesota Fish

Pollutant sources to water are divided into point sources and nonpoint sources. Point sources include any discharge to a water body via a pipe or channel, including publicly owned wastewater treatment plants, industrial waste dischargers, and municipal stormwater discharge. Nonpoint sources, as the name implies, include all other sources, such as the diffuse sources of runoff and air deposition.

In the late 1960s, when mercury contamination of fish was first discovered in Sweden, Canada, Minnesota and other places in the United States, the source of mercury was unknown. It was unclear whether the mercury was derived from local geology or delivered by the atmosphere. Scientists knew that mercury must be methylated before it can be accumulated by fish, but they did not know where methylation occurred, whether it was in soil, water, sediment or in the gut of the fish itself. Nevertheless, without mercury there could be no mercury methylation. In the early 1970s, steps were taken to eliminate the obvious sources of mercury to rivers and lakes. These steps included greatly reducing activities that could cause mercury to be discharged to waterways, such as the use of mercury fungicides in paper making and by mercury cell chlor-alkali plants. But because fish were contaminated in remote lakes that never received any industrial discharge, some scientists suspected that the atmosphere was delivering significant quantities of mercury. Significant geological sources of mercury are localized in cinnabar deposits in such places as the coastal range of California and in Almaden, Spain. The geology of most of the United States, including Minnesota, does not contain concentrated sources of mercury.

Much has been learned about environmental mercury in the decades since. By the early 1990s, it was clear that the atmosphere is the main source of mercury to most surface waters. By 1994, an expert panel (Expert Panel 1994) was able to conclude that anthropogenic activities accounted for 50 to 75% of mercury emissions to the atmosphere, and hence 50 to 75% of mercury deposition to aquatic systems. A more recent study in Minnesota (Engstrom and Swain 1997) indicated anthropogenic sources contribute 70% of the mercury deposition to the state.

The 70% proportion was calculated by the relative change in mercury accumulation in seven rural lakes, as presented in Swain *et al.* (1992). According to those calculations, atmospheric mercury deposition in the rural upper Midwest increased from 3.7 g km^{-2} yr^{-1}) in 1850 to 12.5 g km^{-2} yr^{-1} in 1990. If 3.7 was the natural deposition rate, then natural sources contributed 30% of total deposition in 1990, leaving 70% as anthropogenic. Between 10 and 15 sediment cores from each of seven lakes were needed to account for the total mercury deposition. This type of data collection has not been performed on a national scale.



Ed Swain and Dan Engstrom collecting a sediment core

Our ability to understand and model the atmospheric transport and deposition of mercury is hampered by the various chemical forms that mercury can assume. Mercury can be emitted from anthropogenic (human) sources as either elemental or divalent forms, and most likely can interconvert in the atmosphere through chemical reactions. Essentially all mercury emitted to the atmosphere will eventually deposit on the Earth’s surface. Because of uncertainty about atmospheric chemistry, predicting where or when will deposit is difficult. The fundamental chemical uncertainty is the conversion from elemental mercury vapor to divalent mercury. Elemental mercury vapor is relatively inert, and therefore may remain in the atmosphere up to one year, allowing long-distance dispersal before deposition to terrestrial or aquatic systems. Divalent mercury, on the other hand, is much more readily removed from the atmosphere in rain (“wet” deposition) because it is water soluble and tends to stick to surfaces, including those of airborne dust, vegetation, and even dew (“dry” deposition). Scientists use complex computer models to simulate the atmospheric processes and deposition. The models are continually refined as new scientific understanding becomes available, but because much remains unknown, there is considerable uncertainty in the predictions.

After mercury deposits on to the Earth’s surface only a small proportion of mercury becomes geologically immobilized through burial in the sediments of lakes, oceans or river deltas. Mercury deposited to soils is held for a geologically short time (tens or hundreds of years), from which it can be leached, eroded or volatilized back to the atmosphere. A small proportion of the deposited mercury makes its way to environments where it may be methylated (wetlands, lakes and rivers).

Worldwide trends in mercury emissions and deposition must be in approximate balance, because any emitted mercury must come down somewhere, sometime. Gaseous elemental mercury has an average atmospheric residence time of up to one year, therefore, time lags in deposition should not be significant when viewed on a multi-year basis. Volatilization from soils of atmospherically deposited mercury may produce a significant lag, or smoothing, of deposition compared to primary anthropogenic emissions, but this effect has not been adequately studied and quantified. Erosion or leaching of mercury to aquatic systems from terrestrial soils is known to be significant in some watersheds (e.g., Balogh et al. 1997, Balogh et al. 1998, Balogh et al. 2003), but their effect on time lags also has not been adequately studied.

Despite the difficulty in fully understanding and modeling the chemistry of mercury in the atmosphere, analysis of lake sediments does yield empirical evidence on mercury deposition. Because local geological sources of mercury are relatively rare, mercury in sediments of lakes that never received industrial discharges usually can be attributed to atmospheric deposition. In Minnesota, analysis of sediments accumulated in lakes over the past several hundred years allowed the following conclusions (Swain *et al.* 1992; Engstrom and Swain 1997; Engstrom *et al.* 1999):

- About 70% of current mercury deposition in Minnesota is a result of anthropogenic emissions.
- Annual atmospheric deposition as of 1990 was 12.5 micrograms per square meter per year (total of wet plus dry deposition).

By comparing to sediment cores from coastal Alaska, we can attribute recent (1990s) atmospheric sources to Minnesota as follows: 30% natural from outside the state (e.g., volcanoes), 30% global pollution, and 40% regional pollution (Engstrom and Swain 1997). Several recently published modeling studies (using different models) of global mercury cycling agree with this relative contribution from natural sources (Mason & Sheu 2002, Lamborg 2002, and Seigneur *et al*. 2004).

Atmospheric deposition of mercury in some parts of Minnesota peaked in the 1970s, and has declined slightly since then. Wet deposition rates since 1996 (when mercury deposition network monitoring began) appear relatively uniform around the state, as shown by overlap in annual mercury deposition fluxes from fixed monitoring stations in Minnesota and Brule Wisconsin (Figure 8); the inset bar chart shows the station mean annual fluxes are not significantly different for the period 1996-2003. There is a significant precipitation gradient from northeast to southwest Minnesota, but this does not apparently determine the mercury deposition pattern. The uniformity in deposition indicates that sources causing locally elevated atmospheric deposition have been removed. Likely local sources include poorly controlled waste incineration, mercury fungicide use in paper making, and coal combustion in urban areas for space heating.

Despite removal of many mercury sources, significant mercury emissions sources remain within Minnesota and contribute to mercury deposition in the state. Jackson *et al.* (2000) estimated that one-quarter of the 40% regional pollution, or 10% of total deposition within Minnesota, is because of emissions within the state. The balance of the regional sources was attributed to the rest of the United States.

Engstrom and Swain (1997) apportioned the 70% of atmospheric deposition that is anthropogenic into 30% global and 40% regional, by comparing mercury increases in northern Minnesota to coastal Alaska. By assuming that Alaska is so far from sources that it represents global deposition (which is also a component of Minnesota's deposition), and noting that Minnesota's increase over natural is 3.4-fold, while Alaska's increase is just 2.0-fold, one can calculate that in 1990 the source areas of Minnesota's deposition was 30% natural global, 30% anthropogenic global, and 40% anthropogenic regional.

Unfortunately, there was no way to know the exact geographic extent of the sources contributing to the regionally elevated mercury deposition. Jackson et al. (2000) apportioned the source region as follows, using qualitative knowledge of source strengths and likely transport distances: "…the United States is assumed to be the regional area that Engstrom and Swain determined contributes 40% of the deposition in Minnesota. The 40% is then subdivided: 10% being local deposition from Minnesota emissions; 15%

from the rest of the Midwest and the remaining 15% from U.S. sources outside of the Midwest." The 10% proportion will decline over time as incinerators and other sources of divalent mercury are controlled. We expect that models of later emission years will show a lower and lower in-state percent contribution to overall deposition in Minnesota.

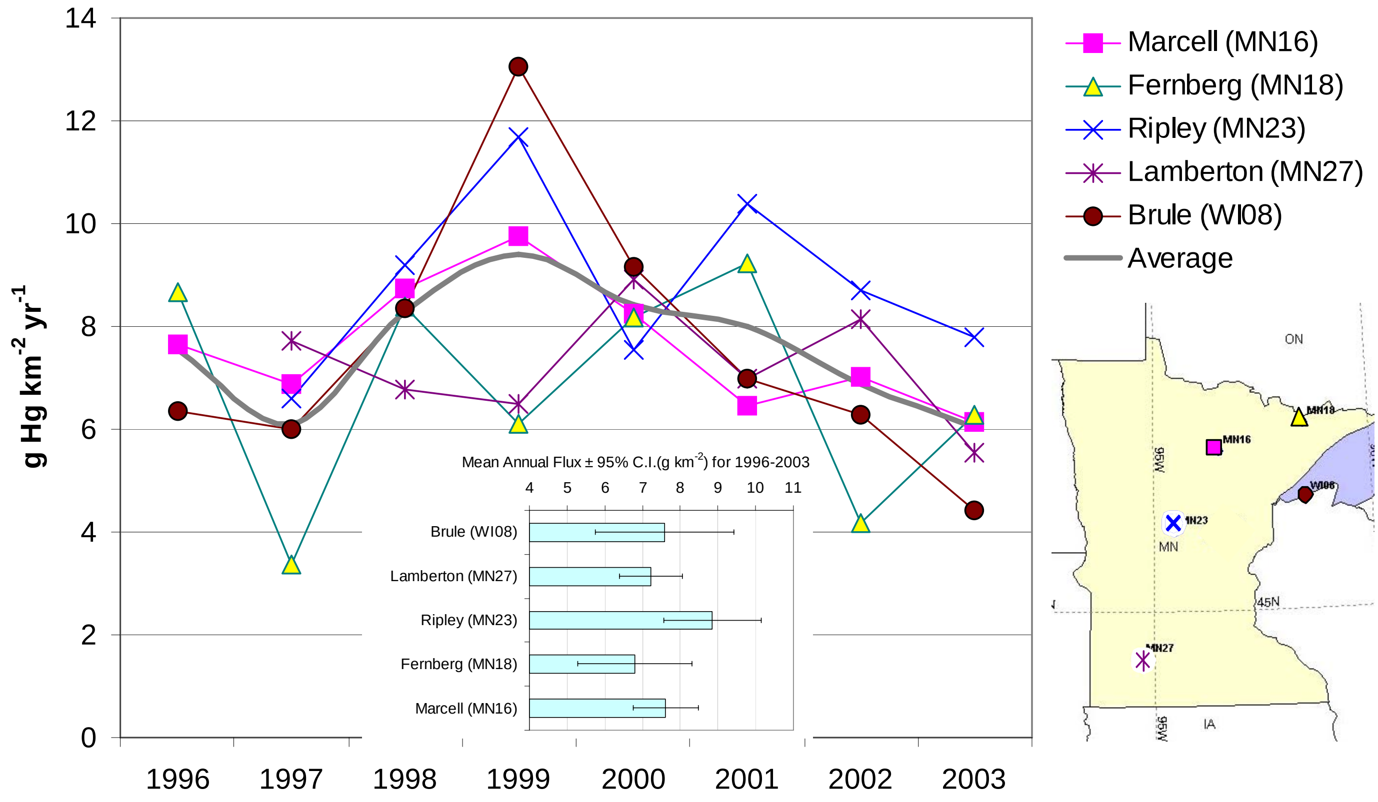


Figure 8 Annual Mercury Flux at Mercury Deposition Network (MDN) Sites in Minnesota

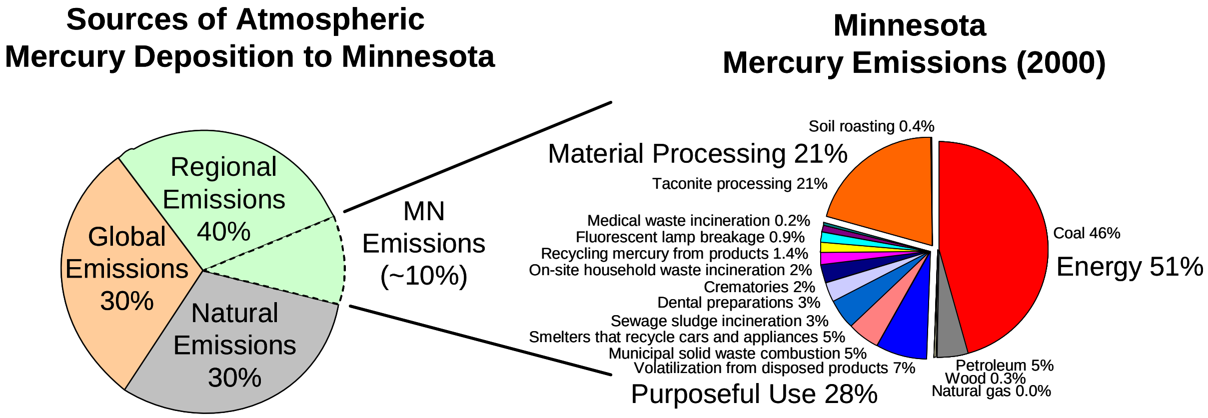
The 10% in-state contribution is supported by atmospheric modeling of mercury for a domain that includes the continental United States, southern Canada, and northern Mexico (Atkinson, 2003). For Minnesota, the USEPA modeling showed total annual emissions of mercury from Minnesota were 2571 kg in 1998, and 586 kg were deposited within the domain. According to model results, of the 586 kg of mercury, 48% (281 kg) was deposited within the state, which is 10.9% of the 2571 kg emitted by the state. Model results showed annual atmospheric mercury deposition in Minnesota was within the range of 10 to 30 g/km^2, which is 2200 to 6600 kg/y for the whole state. The 281 kg deposited by Minnesota sources, therefore, represents 4% to 13% of the total deposition to the state. Thus, the 10% estimated state contribution to mercury deposition within the state, from Jackson *et al.* (2000), is within the bracketed range from USEPA's model results.

One concern when evaluating mercury sources is the likelihood of "depositional hotspots" where mercury deposition far exceeds the norm. Engstrom and Swain (1997) discuss sediment data that show greater mercury loading in 1990 to lakes in urban Minneapolis and agricultural areas of western Minnesota, compared to northern Minnesota (findings confirmed by later unpublished sediment core work). However, it is uncertain if the greater loading is delivered by the atmosphere. In agricultural areas, the mercury load to lakes is likely enhanced by soil erosion. In urbanized areas, the mercury load may have been enhanced by development of impervious surfaces or indirectly from the use of mercury-preserved latex paints. Regardless of the indication of some historical localized higher mercury loading rates, there is no evidence of subsequently higher fish mercury levels (i.e., no biological hotspots).

An alternative to sediment core data as the basis for total mercury deposition is computer modeling. There has been no effort to correlate Minnesota's fish concentrations to any atmospheric modeling, such as that of Atkinson (2003). There is a great deal of variation in fish concentrations from lake to lake, even in rural areas far from emission sources that receive equal atmospheric mercury deposition. We know that the average mercury concentrations of fish in the Minneapolis-St. Paul metropolitan area are not significantly different from concentrations in the rest of the SW region. Even though lake to lake variation is greater than any increment because of urban deposition, our working assumption is that the mercury concentrations of fish in all lakes will decline when a steady state is approached after the atmospheric loading is reduced. The EPA model output (Atkinson, 2003) shows greater deposition near Minneapolis for the 1998 emission year. The greater deposition was driven by two incinerators in Goodhue County, a rural area over 40 miles southeast of Minneapolis. By 2000, mercury emissions from these facilities were reduced from about 300 pounds to less than 5 pounds per year.

In the 1990s there were three incinerators operating in Goodhue County, one in Cannon Falls, and two in Red Wing. Lake and stream data for mercury are sparse in the Goodhue County area, largely because there is not much surface water aside from the Mississippi River and some tributaries. One tributary, the Cannon River, flows in an easterly direction in Rice County and then flows into Goodhue County, near the northern county line. The Cannon River discharges into the Mississippi about 5 km NE of the city of Red Wing, the site of the other contributing incinerator. The headwater reaches in Rice County are impaired, and listed for excess mercury in fish. In addition Byllesby Reservoir is listed as impaired because of fish that were sampled in the 1980s. Byllesby Reservoir is adjacent to the city of Cannon Falls, the location of one of the two incinerators that contributed to the modeled elevated mercury deposition. However, the incinerator did not start operating until the 1990s, after the fish were sampled. The Cannon River between Byllesby Reservoir and Welch, MN was sampled in 1992 and the fish (carp, smallmouth bass, and walleye) were not impaired and these reaches are not listed for excess mercury.

One basic tenet of the TMDL program is to make the best TMDL possible with the data available. Our available fish data, albeit sparse in some locations, do not support the presence of a hotspot in terms of fish mercury concentrations in Goodhue County. Once the reduction goals are established, a phased adaptive watershed management approach will re-assess subsequent data as needed.


Sources of Atmospheric Mercury Deposition to Minnesota

Minnesota Mercury Emissions (2000)

Figure 9 Sources of Mercury Deposition and Estimated Mercury Emission Sources in Minnesota

Mercury from anthropogenic emission sources (i.e., emitted to the atmosphere because of human activities) is divided by the MPCA into three categories: (1) emissions related to energy production, (2) emissions because of purposeful use, and (3) emissions because of material processing (Figure 9). Although emissions from fossil fuel combustion and the processing of metal ores are both the result of the incidental release of trace contaminants of natural geological materials, we have placed them in separate categories (energy production and material processing, respectively). As of 2000, 51% of Minnesota's mercury emissions are from energy sources, 21% from taconite processing, and 28% from purposeful uses.

5.2 Principle of Proportionality for Mercury in Air and Biota

Ideally, the link between emissions and mercury bioaccumulation in fish would be known quantitatively and the effect of a given reduction in emissions accurately modeled. Such models are under development. In the absence of a validated model that accurately incorporates the complexities of atmospheric chemistry, watershed transport, methylation, and bioaccumulation in fish; we rely on the following rationale (Jackson *et al*. 2000):

a. A reduction in emissions from sources in a given source area (local, regional or global) results in a proportional reduction in the rate of deposition in Minnesota attributable to those sources.

b. A reduction in deposition results in a proportional reduction in mercury loading to water bodies.

c. Within a given water body, a proportional reduction in mercury loading in the water results in a proportional reduction in mercury concentrations in fish.

For example, a 50% reduction in mercury emissions from Minnesota sources is projected to result in a 5% reduction in deposition in Minnesota (50% of 10% state share), and, therefore, a 5% reduction in mercury contamination of fish. A 50% reduction applied to anthropogenic global sources is projected to result in a 35% reduction in deposition in Minnesota (50% of 70%), and, therefore, a 35% reduction in mercury contamination of fish.

This assumption of proportionality between mercury deposition and fish bioaccumulation acknowledges that lakes differ in the proportion of atmospherically deposited mercury that is methylated and the efficiency that methylmercury is accumulated in fish. This is described below as the bioavailability factor. Proportionality assumes that these factors are constants, unaffected by the rate of atmospheric mercury deposition.

A variety of physical processes could cause time lags, including delayed release of mercury from the terrestrial watershed around surface water, and release from sediments contaminated from earlier atmospheric deposition. However, mercury in terrestrial systems will eventually reach a new steady state with atmospheric deposition, and total loading of mercury to surface water will be proportional to atmospheric deposition.

Similarly, sediments eventually will also equilibrate with atmospheric deposition, but perhaps through different mechanisms than do terrestrial soils. For example, if the sediment is in a depositional zone, new, cleaner sediments may simply cover up and bury the older, more contaminated sediment.

Mechanistic models for mercury predict proportionality between atmospheric deposition and fish contamination. The Pilot Mercury TMDL exercise for the Florida Everglades found “The E-MCM model predicts a linear relationship between atmospheric mercury deposition and mercury concentrations in largemouth bass….The slight offset from a 1:1 relationship results from slow mobilization of historically deposited mercury from deeper sediment layers to the water column….” (Atkeson *et al.* 2002; Figure 10).

Similarly, the EPA’s Mercury Maps project examined the MCM model and the IEM-2M watershed model, and found they both predict proportionality between atmospheric deposition and fish contamination (USEPA 2001d). The following approach follows the Mercury Maps discussion on the proportionality between change in atmospheric deposition and change in fish tissue concentration. The Mercury Maps report describes the relationship as

$$\frac{C_{fish,t2}}{C_{fish,t1}} = \frac{\left(L_{air,t2} + L_{other,t2}\right)}{\left(L_{air,t1} + L_{other,t1}\right)} \tag{1}$$

where, $C_{fish,t1}$ and $C_{fish,t2}$ are the mercury concentrations in fish at times 1 and 2, which could be the baseline and target concentrations; $L_{air,t1}$ and $L_{air,t2}$ are the air deposition Hg loads to a waterbody, including direct deposition and indirect deposition via the watershed; and L_{other} is loading from other sources. There are no known significant natural geologic sources of mercury in the state. For this mercury TMDL, the only other sources considered are wastewater discharges (NPDES permitted discharges). The wastewater sources are insignificant compared to air sources (See Section 6.3) and, therefore, drop out of the simplified equation.

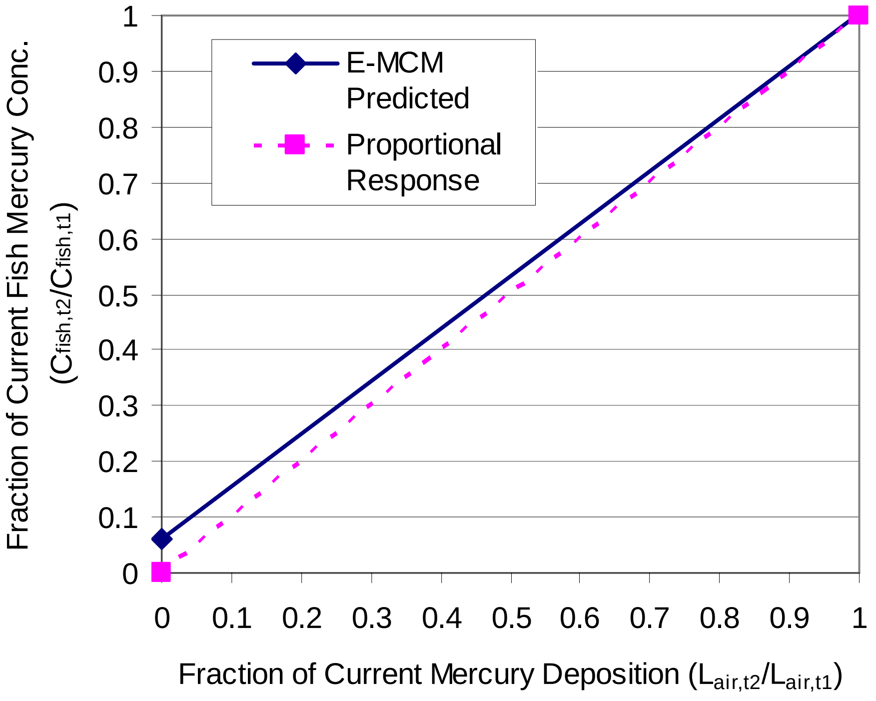


Figure 10 Proportionality between atmospheric mercury deposition and mercury concentrations as predicted by the E-MCM model for largemouth bass in the Everglades

Atmospheric loading is the product of area and air deposition; total area can be split into water area and land area to distinguish direct atmospheric loading from indirect watershed loading. To account for mercury that is buried in the soil or volatilized to the atmosphere, the watershed loading can be discounted by a runoff coefficient, which remains constant for a given region as long as there are no significant changes in land cover/use. This was tested by comparing land cover changes between 1982 and 1997 (http://www.mn.nrcs.usda.gov/technical/nri/tables/lcu_change.htm), applying standard runoff coefficients to each of the general land cover types. Although there were obvious increases in urban land use, the effect of the change was not significant to the composite runoff coefficient for the state: composite runoff coefficients were 0.289 for 1982 and 0.287 for 1997.

The air deposition mercury loading can be described as

$$L_{air} = D_y \cdot \left(A_L \cdot r + A_W\right) \qquad (2)$$

where D_y is the annual air deposition flux of mercury (g km^{-2} y^{-1}); r is the runoff coefficient (also known as the delivery ratio); A_L and A_W are the areas of land and water (km^2). Assuming areas and r for each region do not change from t_1 to t_2, Substituting this definition of L_{air} into equation 1, areas will not change from t_1 to t_2 and, therefore areas drop out of the equation. In addition, we can include a bioavailability factor to account for the fraction of divalent mercury (Hg^{2+}) converted to methylmercury (MeHg). Combining Equations 1 and 2, as well as, including the bioavailability factor, the relationship becomes

$$\frac{C_{fish,t2}}{C_{fish,t1}} = \frac{D_{y,t2}}{D_{y,t1}} \cdot \frac{r_{t2}}{r_{t1}} \cdot \frac{b_{t2}}{b_{t1}} \qquad (3)$$

where *b* is the bioavailability factor. An operational definition for *b* is the MeHg fraction of total Hg, because MeHg is the form of mercury that bioaccumulates; however, it could also include labile Hg^{2+} that is readily converted to MeHg. Bioavailability is influenced by a variety of factors that control methylation and MeHg mobility, such as wetland density, sulfate deposition, and dissolved organic carbon production.

For the estimate of the TMDL for mercury, we are assuming *r* and *b* do not change over time; therefore, their t2/t1 ratios are equal to one, they can drop out of the equation, and Equation 3 simplifies to

$$\frac{C_{fish,t2}}{C_{fish,t1}} = \frac{D_{y,t2}}{D_{y,t1}} \qquad (4)$$

We can rearrange Equation 4 to solve for a target atmospheric deposition (D_y), substituting the baseline and target values for the t_1 and t_2 values, respectively:

$$D_{y,target} = \frac{C_{fish,target}}{C_{fish,baseline}} \cdot D_{y,baseline} \qquad (5)$$

To calculate a target atmospheric mercury load, we are not concerned with the values of *r* and *b* because we assumed they do not change over time; however, it is plausible that *r* and *b* could change. We can consider what affect changing these coefficients will have on achieving the target fish tissue concentration. The combined effect of *r* and *b* determines how much Hg is available for uptake by the aquatic food chain. We expect *r* to be greater in the SW region than in the NE region and the converse for *b*. In other words, in the SW waters, such as the Minnesota River, we expect a high transport rate of Hg off the watershed, but a small fraction of that runoff Hg is bioavailable; whereas, in the NE, we expect a low transport rate, but a high fraction of bioavailable Hg.

Neither proportionality, nor any other model, is able to deal accurately with short-term perturbations in equilibrium, such as accelerated erosion of soil that contains mercury from past atmospheric deposition. It is not clear, for instance, whether the mercury associated with eroded soil is just as likely to become methylated as other sources of mercury. New research suggests that more recently deposited mercury is more bioavailable (Hintelman et al. 2002). Nevertheless, intentional reduction of *r* through watershed best management practices could contribute to load reductions. This is reasonable for the SW region, where we have evidence of Hg associated with suspended solids, but not the NE region, where Hg is more often associated with dissolved solids (i.e., dissolved organic matter). On the other hand, fish mercury concentrations in the NE region could be reduced by reducing *b* via reduced air deposition of sulfate, which can reduce methylation rates by reducing the activity of sulfate reducing bacteria (See Section 5.4). Reducing sulfate deposition is not likely to have an effect in the SW region because sulfate concentrations are relatively very high and wetland acreage has significantly decreased over the last 100 years.

Although there is little research on the interaction of *r* and *b*, it is likely that in SW Minnesota, *r* and *b* are not entirely independent of each other. For example, when *r* is high, soil erosion delivers suspended solids and phosphorus, as well as mercury, to the surface water. These materials probably reduce bioavailability (*b*) of Hg. The suspended solids sorb divalent mercury, perhaps reducing interaction with methylating bacteria. Greater phosphorus increases the biomass of all levels of the food chain, decreasing the concentration of methylmercury in fish (i.e., biodilution).

We can also consider a change in the loading from the "other" sources. Point sources are treated as insignificant in the loading calculations, but they could be included in the equation for the sake of discussing potential reductions in point source loads. Point source loads are the product of discharge flow and mercury concentration. It is likely that the average mercury concentrations in many wastewaters will be reduced in the future through changes in dental practices and improved wastewater treatment methods, such as "Bio-P" and "Chem-P," which reduce mercury concentrations in the effluent by reducing solids (G. Kimball, MPCA; personal communication).

5.3 Trends in Mercury Emissions and Deposition

5.3.1 Trends in Use and Emission of Mercury

While some of the mercury used in products is recycled and reused, some is released into the environment when it is spilled or when it is disposed of in a drain or garbage. Historically, many intentional uses presumed mercury dissipated into the environment and was not harmful (e.g,. fungicides: seed coatings, golf course treatments, latex paint preservatives). As understanding about the bioaccumulation of mercury in the aquatic food chain grew, these uses of mercury were banned.

Mercury use peaked in the 1960s in the United States at more than 2,000 metric tons per year and by 1990 declined significantly to about 600 metric tons (Figure 11A; Engstrom and Swain 1997). Worldwide consumption followed this same pattern ((Hylander and Meili 2003). U.S. industrial consumption of mercury has consistently been about 25% of global production of mercury (Engstrom and Swain 1997). As noted above, only a portion of the mercury consumed in manufacturing and products is emitted to the atmosphere. However, mining and the use of mercury was historically conducted on such a massive scale that if only 10 to 15% of this mercury were emitted, it would easily exceed mercury emissions from other sources. In Figure 11A one can see that the mercury used as fungicides in agriculture, paper, and paint totaled about 400 metric tons in 1970, and presumably all volatilized to the atmosphere within a few years. Compare that total to the mercury content of coal (Figure 11B), which approaches 80 metric tons in the 1990s, of which about 50 tons were emitted (USEPA 1997). Given the significant decline in mercury use, one would expect mercury emissions to have declined significantly in recent years.

Future mercury emissions from manufacturing and fuel combustion are expected to decline worldwide. Pirrone *et al.* (1996) concluded that on a global basis, total anthropogenic emissions of mercury peaked in 1989 at about 2,290 metric tons, and have been decreasing at the rate of about 1.3% per year. Use of mercury in manufacturing and products will continue to decline world wide, resulting in lower emissions. Mercury emissions associated with coal combustion are also expected to eventually decline worldwide; however, growth in Asia may significantly delay the expected reductions.

A historical reconstruction of emissions in the United States from 1930 to 2000 (Figure 12; Pollman and Porcella 2002) shows a significant decline in total mercury emissions since about 1980, accelerating after 1990 because of the elimination of mercury from latex paint. The pattern was also seen in mercury emissions from 1870 to 2000 in Maritime Canada (Sunderland and Chmura 2000). Therefore, the amounts of mercury used and released in Minnesota, the United States and the rest of the world have undoubtedly been cut dramatically over the past few decades.

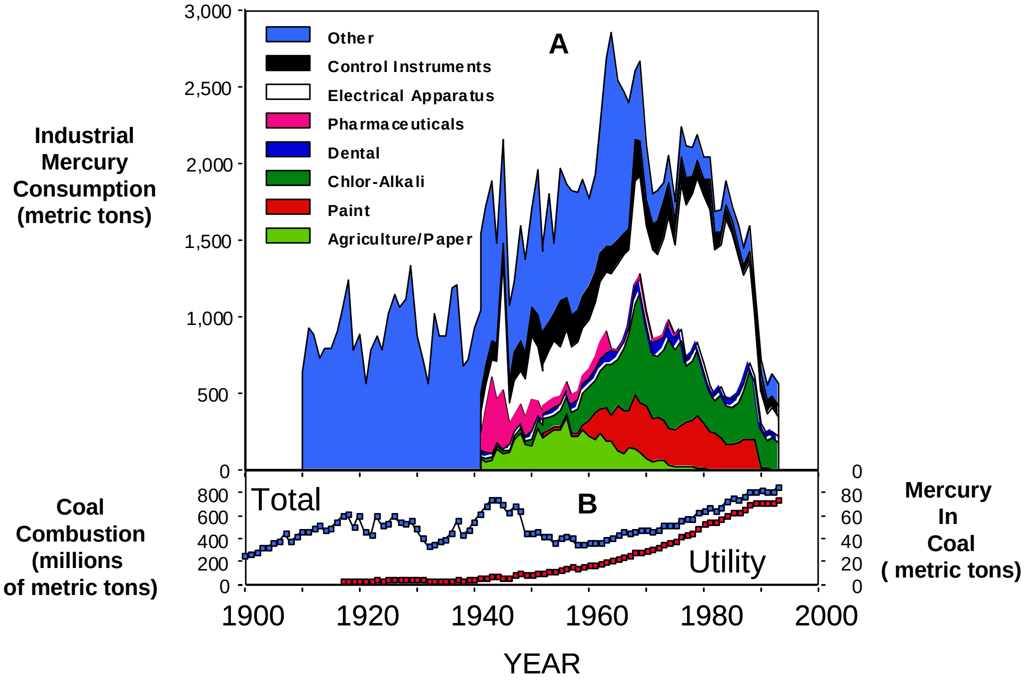


Figure 11 Mercury Consumption in the United States

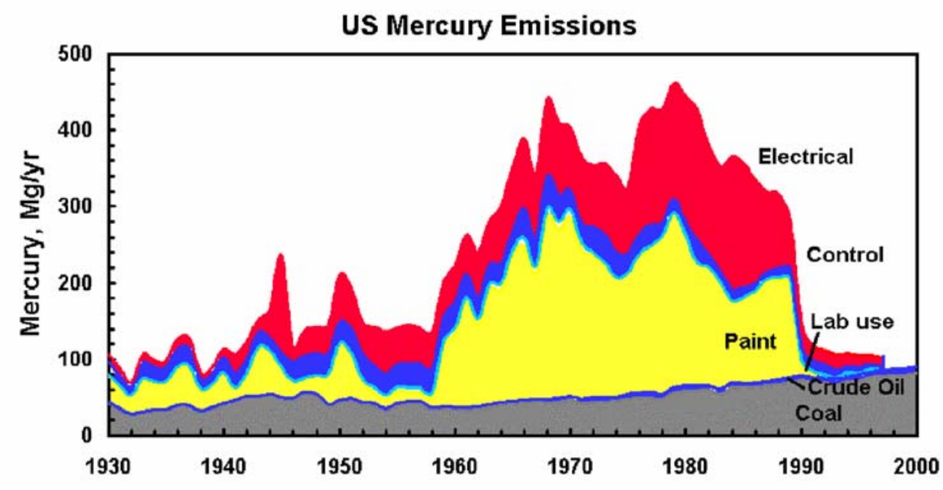


Figure 12 United States Mercury Emissions by Category

Annual mercury releases, as of 2000, in Minnesota are about one-third what they were in 1990 (See MPCA 2005). Current MPCA estimates indicate that statewide releases in 2000 were about 1,650 kg (3,638 lb), 68% below estimated 1990 levels (Figure 13). Nearly all reductions since 1990 are because of product-related uses: restricting the intentional use of mercury in products such as paint and batteries, improved management and substitution of mercury-containing products (thermostats, fluorescent lamps, etc.), reduction in uncontrolled incineration, and emission controls on waste-combustion facilities.

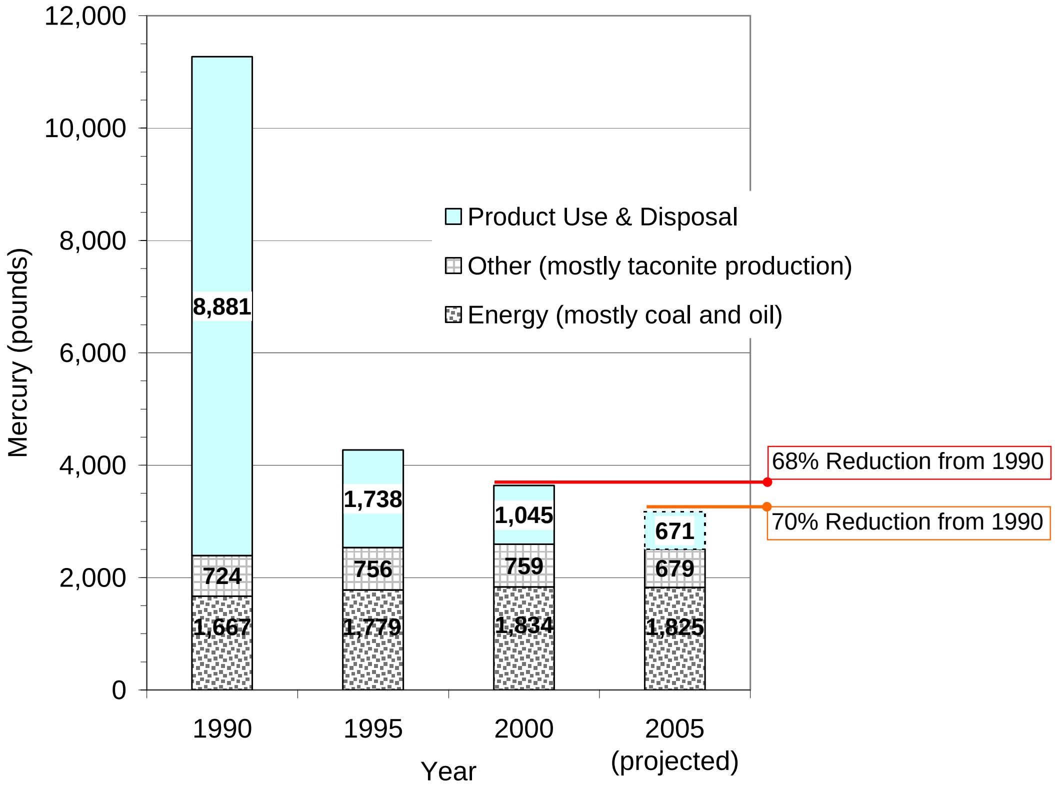
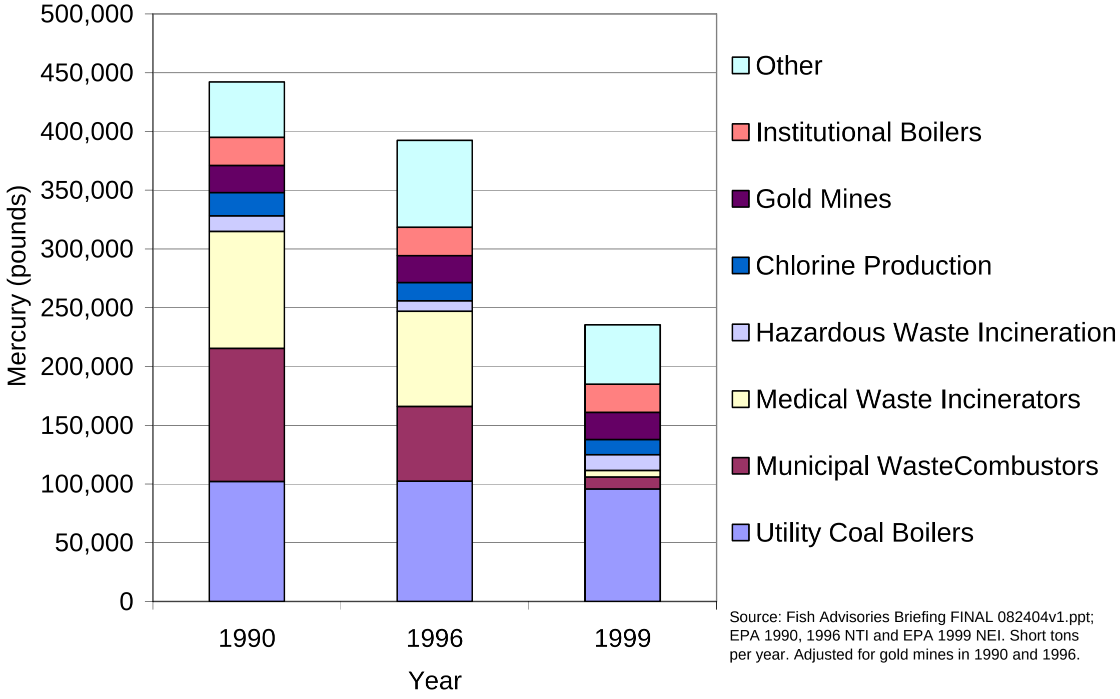


Figure 13 Minnesota Statewide Mercury Emissions Trend by Source

Given existing trends in decreased use of mercury in products, increases in mercury-product recycling and controls on sewage-sludge incinerators, the MPCA expects emissions to continue to decline, falling below 70% of 1990 levels by 2005. Taconite data shows an increase of 4.8% from 1990 to 2000, and energy production shows an increase of 10.0% over that time period. Because of an 88% decrease in mercury emissions from Product Use and Disposal, a much bigger category, there was an overall decrease of 68% in Minnesota Hg emissions from 1990 to 2000. Because of this large overall decrease, and because both of these sectors mostly emit elemental mercury (about 90% elemental), we do not expect any noticeable effect of these modest increases on local deposition or mercury concentrations in fish. These increased emissions are calculated into the state's overall trend since 1990, and do not affect the establishment of the 1990 baseline. This information will, however, be a key component of subsequent implementation planning.

Anthropogenic emissions in the U.S. have decreased 45% between 1990 and 1999, according to the USEPA (Figure 14). This mercury emissions summary does not include area sources, such as latex paint and fungicides, which are included in the Minnesota Mercury Emissions Inventory (MPCA 2005). Before the USEPA banned the use of mercury in interior paint in 1990 and exterior paint in 1991, the paint sector was using 150-350 tonnes per year. Mercury was estimated to volatilize from paint at the rate of 75% per year (Barr Engineering 2001). In 1989, 211 tonnes (465,170 pounds) of mercury were used by the U.S. paint industry. Thus, if emissions from paint were included in the U.S. emissions summary, the 1990 emissions estimate would be much larger and the decline in mercury emissions would be much larger than the 45% decline show in Figure 14 (USEPA 2004).



Source: Fish Advisories Briefing FINAL 082404v1.ppt; EPA 1990, 1996 NTI and EPA 1999 NEI. Short tons per year. Adjusted for gold mines in 1990 and 1996.

Figure 14 USEPA's anthropogenic mercury emissions in U.S.

5.3.2 Trends in Atmospheric Deposition of Mercury

A number of lines of evidence support the conclusion that atmospheric mercury has declined because of decreased mercury use and emissions. An analysis of air concentrations from around the world since the 1970s shows significant decreases of about 20% since about 1990 (Slemr *et al*. 2003).

Lichens and mosses have been used to monitor atmospheric mercury concentrations. Utilizing lichens in a national park in North Dakota as a natural collector of atmospherically deposited metals, Bennett and Wetmore (2000) found that mercury concentrations decreased by about 30% from 1983 to 1998. Based on nationwide surveys of moss as a collector of atmospheric deposition of metals, Steinnes *et al.* (2003) showed that mercury concentrations in 2000 were 30% lower than they had been in 1985.

Evidence of declines from contemporaneous measurement of mercury in precipitation is more problematical because mercury deposition is highly dependent on the quantity of precipitation, which varies significantly from year to year. The statistical effect of precipitation variability should become less once a few decades of data are collected. Unfortunately, a permanent, long-term national monitoring program — the Mercury Deposition Network (http://nadp.sws.uiuc.edu/mdn/) — was not begun until 1995. In a monitoring program funded by the State of Minnesota, Glass and Sorensen (1999) saw slight increases in mercury deposition in the Upper Midwest from 1990 to1995. Between 1994 and 1999, with a different monitoring program, Watras *et al*. (2000) reported a 40% decrease in mercury deposition in northern Wisconsin.

The initial evidence that mercury deposition rates had decreased over the past few decades came from sediment cores from a number of lakes in Minnesota (Engstrom and Swain 1997), although there was not yet evidence for global declines. Sediment core studies from lakes in Minnesota and elsewhere show slight declines in atmospheric deposition relative to a peak in the 1970s and 1980s. There is some evidence that concentrations of mercury in fish have also declined, but not to the point of eliminating concerns about fish consumption (see Section 5.6). But, it is encouraging that efforts to reduce the use and release of mercury appear to have resulted in measurable environmental improvement.

Further evidence for declines in midcontinental North America comes from the analysis of an ice core from the Fremont Glacier in the Wind River Range, Wyoming (Figure 15; Schuster *et al.* 2002). From this historical trend reconstruction, one can see the approximate magnitude of the pre-industrial (or natural) background level of mercury cycling through the environment, and spikes in mercury caused by events such as volcanoes. The ice core record shows a distinct decline since the 1980s. Evidence of declines in atmospheric deposition of mercury in New England has been found from bog cores in Maine (Norton *et al.* 1997) and multiple lake cores in Vermont and New Hampshire (Kamman and Engstrom 2002).

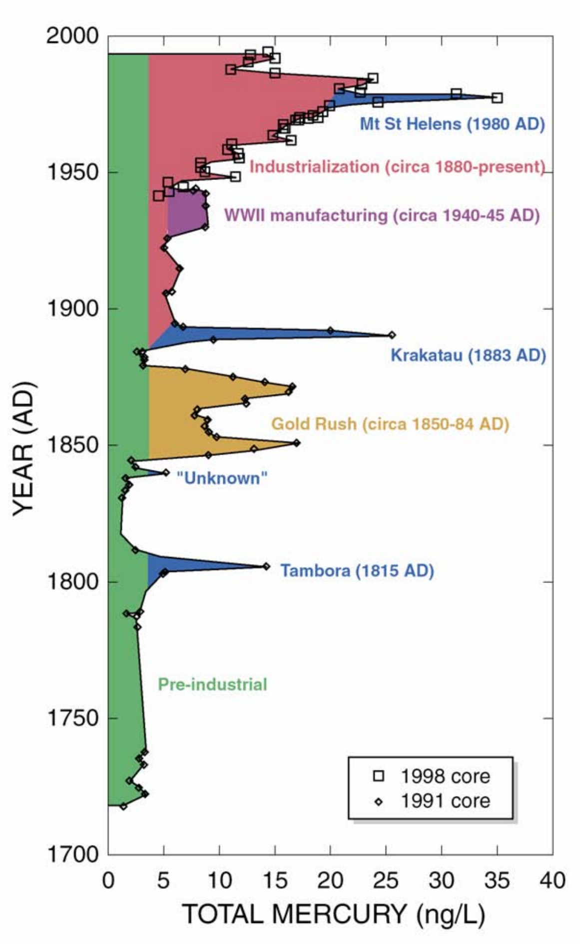


Figure 15 Mercury Accumulation Record in the Fremont Glacier, Wyoming

5.4 Interaction Between Sulfate Deposition and Mercury Methylation

Mercury contamination of fish is expected to be proportional to atmospheric deposition of mercury if the proportion of mercury that is methylated is constant. However, it is likely that the proportion has increased, relative to natural levels, because of other anthropogenic changes. Sulfate-reducing bacteria (SRB) have been shown to be responsible for most of the transformation of deposited mercury into methylmercury. Atmospheric deposition of sulfate is thought to have stimulated the activity of SRB in geographic areas that are naturally sulfate-poor and, therefore, have increased the proportion of mercury that is methylated (Gilmour and Henry 1991, Gilmour et al. 1992, Branfireun *et al.* 1999, Jeremiason *et al.* 2003, Jeremiason *et al.* 2006).

Therefore, it is possible that decreases in sulfate deposition as a result of the Clean Air Act of 1990 may decrease the efficiency of mercury methylation. Accordingly, we would expect two synergistic forces working to decrease mercury concentrations in fish: (1) decreased mercury deposition and (2) decreased activity of the sulfate-reducing bacteria that methylate mercury. This synergism is a margin of safety because it is not factored in to the TMDL calculations

As noted above, northern Wisconsin has seen a 40% decrease in mercury deposition. In a more recent study, Hrabik and Watras (2002) concluded that mercury in fish from Little Rock Lake (in northern Wisconsin) decreased by roughly 30% between 1994 and 2000 because of decreased atmospheric mercury deposition, and 5% because of decreased atmospheric sulfate deposition (30% in a basin that had been isolated and acidified as an experiment). An ongoing study in northern Minnesota includes adding sulfate to a wetland and the response has been increased methylmercury concentrations and export from the wetland (Jeremiason *et al*. 2003, Jeremiason *et al.* 2006).

5.5 Point Sources to Water

There are 580 NPDES permitted wastewater treatment plants (WWTPs) that discharge to Minnesota's mercury impaired waters; 270 of those WWTPs discharge to receiving waters covered under this statewide mercury TMDL: 203 in the SW region and 67 in the NE region (Appendix B). Most of the dischargers are publicly owned treatment works (POTWs). In addition, the following facilities were included in the point source mercury load estimates: NE region has one electricity-generating coal-fired power plant, four pulp & paper mills, and seven taconite processing facilities; the SW region has two electricity-generating coal-fired power plants and one petroleum refinery. Other major NPDES discharges are cooling water, which do not contain added mercury and were, therefore, not included in the point source mercury load estimates.

Wastewater is comprised of source water and anthropogenic wastewater inputs. Essentially all the mercury (99%) in WWTPs is assumed to be added to the waste stream and a conservatively estimated 1% is from drinking water. USEPA (2001d) reported the Association of Metropolitan Sewerage Agencies (AMSA) had done a study of 24 POTWs in six states, using clean techniques for sampling and analysis that reported a median mercury concentration of 5.0 ng/L in POTW effluents. MPCA has requested similar monitoring from 37 NPDES facilities (POTWs and industrial) in Minnesota; the central tendency of mercury concentrations in effluent has been in the range of 4 to 6 ng/L; therefore, 5 ng/L was used as the typical mercury concentration for NPDES facilities in Minnesota.

Stormwater is considered wastewater as well, and is comprised of source water (atmospheric deposition) and direct anthropogenic inputs (wastewater). Unlike other wastewater, stormwater source water is precipitation runoff. As such, the relative percentages of background source and direct anthropogenic sources are approximately opposite the WWTP influent proportions and essentially all mercury in stormwater is attributed to source water (See Section 6.2). The Western Lake Superior Sanitary District has been doing detailed evaluations of mercury influents for a decade and stormwater has not been identified as a mercury source to the POTW (WLSSD 1997).

The MPCA compared mercury concentrations in runoff from cultivated land and urban land use to see if the latter had higher mercury concentrations, which would indicate a significant direct anthropogenic source in urban areas that is not seen in rural areas. Measurements of mercury concentrations in runoff from agricultural field snowmelt and urban stormwater runoff are very limited. In 1996 and 1997, two studies measured snowmelt runoff from agriculture fields (Balogh *et al.* 2000; Sloan *et al.* 2001). In 2002 and 2003, the Minneapolis Park and Recreation Board (MPRB) conducted a pilot study of mercury in stormwater at two locations (one residential and one industrial) for the Minneapolis-St. Paul NPDES Municipal Stormwater Permit. Mercury and total suspended solids grab samples were collected at the two sites during storm events. The median mercury concentration for agriculture runoff was 19.1 ng/L and the median for the urban runoff (i.e., stormwater) was 21.7 ng/L. Considering that the measured agriculture runoff was only snowmelt, whereas the urban runoff data included storms throughout the summer, more overlap in agriculture and urban mercury concentrations would be expected if the agriculture runoff included runoff from summer storm events. These results indicate there is no significant difference between agricultural and urban stormwater runoff; therefore, the additional mercury contribution to stormwater from sources other than atmospheric deposition is essentially zero. In other words, wet and dry deposition account for all of the mercury observed in stormwater and there are no significant direct anthropogenic sources of mercury added to the stormwater.

MPCA also has data for mercury concentrations in taconite processing facilities effluents. Based on the state's NPDES discharge monitoring database, a typical mercury concentration for effluent from these facilities is 1.5 ng/L. USEPA (2001d) reported average mercury concentrations in effluent from pulp & paper mills of 13 ng/L; therefore, this value was used in mercury loading estimates in 1990 because insufficient information for Minnesota mills. Recent monitoring from Boise Cascade had average mercury concentrations of 1.6 ng/L and data collected from Wisconsin paper mills showed an average effluent mercury concentration of 2 ng/L (Mugan 2005).

The mercury contribution from a point source may be insignificant on a state, regional, or even watershed scale, but there would remain the issue of the impact of the discharge on the immediate receiving water. Although theoretically an issue in Minnesota, there is no documented case of locally elevated fish contamination because of a point-source water discharge. Minnesota is building a database of mercury concentrations in wastewater effluent by relying on the wastewater treatment plant operators to routinely collect samples for mercury using clean techniques and low-level mercury analysis.

5.6 Environmental Trends

5.6.1 Summary of Scientific Information

Mercury contamination of fish in almost all Minnesota surface waters will ultimately reach a steady state with atmospheric deposition of mercury. Therefore, changes in anthropogenic emissions of atmospheric mercury that are deposited in Minnesota control changes in fish concentration. Sources of atmospheric mercury deposition to Minnesota include natural (about 30% of total) and local, regional and global anthropogenic sources (totaling the remaining 70%). To the extent that emissions from these three geographic categories are reduced, fish contamination in Minnesota will also be reduced, although, (a) reductions in sources farther from Minnesota will have correspondingly less benefit and (b) lag times of unknown duration will occur because of delayed release of mercury from terrestrial systems and release from contaminated sediments.

Ed Swain adjusting experimental sulfate addition to a wetland

According to emission-inventory reports, emissions of mercury have been significantly reduced on all three geographic scales (i.e., local, regional, and global). Supporting evidence includes changes in the mercury concentrations in sediment, ice cores, moss and lichens, and changes in air concentrations from remote areas of the globe (See Section 5.1).

In addition, reduced sulfate deposition because of mandated controls sulfur dioxide emissions is expected to magnify the benefit of reduced mercury deposition in many lakes in Minnesota and elsewhere. Sulfate deposition can stimulate the activity of sulfate-reducing bacteria (SRB), which are responsible for the transformation of mercury into methylmercury, the form that bioaccumulates in fish (See Section 5.4).

5.6.2 Predicting Response Time

The Everglades ecosystem differs significantly from aquatic systems in Minnesota, but the principle of proportionality between atmospheric mercury deposition and fish mercury concentrations is expected to hold in any ecosystem that does not have local geological sources. In the Everglades, the E-MCM model predicted slow recovery after reductions in atmospheric deposition because macrophyte roots mobilized historically deposited mercury from sediment as deep as 48 cm. Atkeson *et al*. (2002) found that even after 200 years of model simulation, deep sediment layers had not fully adjusted to changes in mercury loadings in the system, and continued to supply Hg(II) and methylmercury to the overlying system. After running simulations for 200 years with current loads to approximate steady state, they reduced atmospheric deposition as a step function and continued the simulation for an additional 200 years. Modeling results indicated the time required to achieve 50% of the ultimate response is approximately 10 years (Figure 15; Atkeson *et al.* 2002); 90% of the ultimate response is projected to occur within 30 years. Projections would, of course, vary for other ecosystems, but this modeling exercise does give an idea of the response time following reductions in atmospheric mercury deposition.

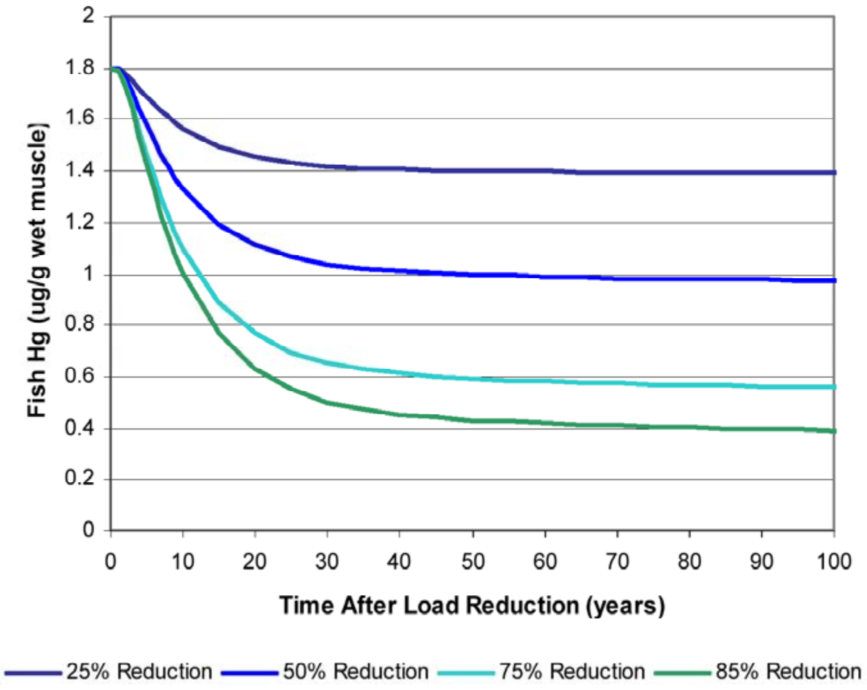


Figure 16 Predicted Dynamic Responses of Fish Tissue Mercury Concentrations Following Reductions in Mercury Deposition

5.6.3 Trends in Fish Contamination

Declines in mercury emission and deposition should result in reduced mercury concentrations in fish. There is evidence of lowered fish contamination levels from Minnesota, Wisconsin (Hrabik and Watras 2002) and Florida (Atkeson *et al.* 2002). Mercury in fish is difficult to monitor and communicate as a uniform measurement because concentrations vary by species and size. Nevertheless, it is possible to monitor temporal trends in fish contamination by monitoring a single fish species, and, within that species, normalizing to a standard length. For example, in Little Rock Lake, Wisconsin, Hrabik and Watras (2002) documented that mercury in fish decreased by roughly 30% between 1994 and 2000, mostly because of decreased atmospheric mercury deposition and partly because of decreased sulfate deposition. There is also evidence that mercury concentrations in largemouth bass in the Everglades have declined in response to lower mercury deposition (Atkeson, pers. com.).

In Minnesota, spatial and temporal trends in fish mercury concentrations have been examined using the fish tissue concentration in standard size northern pike and walleye. Results discussed in Section 4.3 demonstrated significantly higher mercury concentrations in northern pike and walleye in the NE region compared to the SW region. Lakes in the NE region tend to have lower pH, lower conductivity and higher color (i.e., dissolved organic carbon).

Another approach to looking at temporal trends in standardized fish tissue mercury concentrations has been on an individual lake basis. Changes over time in fish mercury concentrations within lakes were evaluated by comparing a recent sample year to an earlier year, which were at least five years apart. Recent sample years were 1995 or later and at least three fish (or composites) were analyzed each year. Of the 176 lakes meeting these criteria, 87 lakes (49%) showed a decrease in mercury concentrations, 45 lakes (26%) had increased fish-Hg, and 44 lakes did not show a significant difference between years (Figure 17). A Chi-square statistical test shows that significantly more lakes declined in fish contamination than increased (p<0.01).

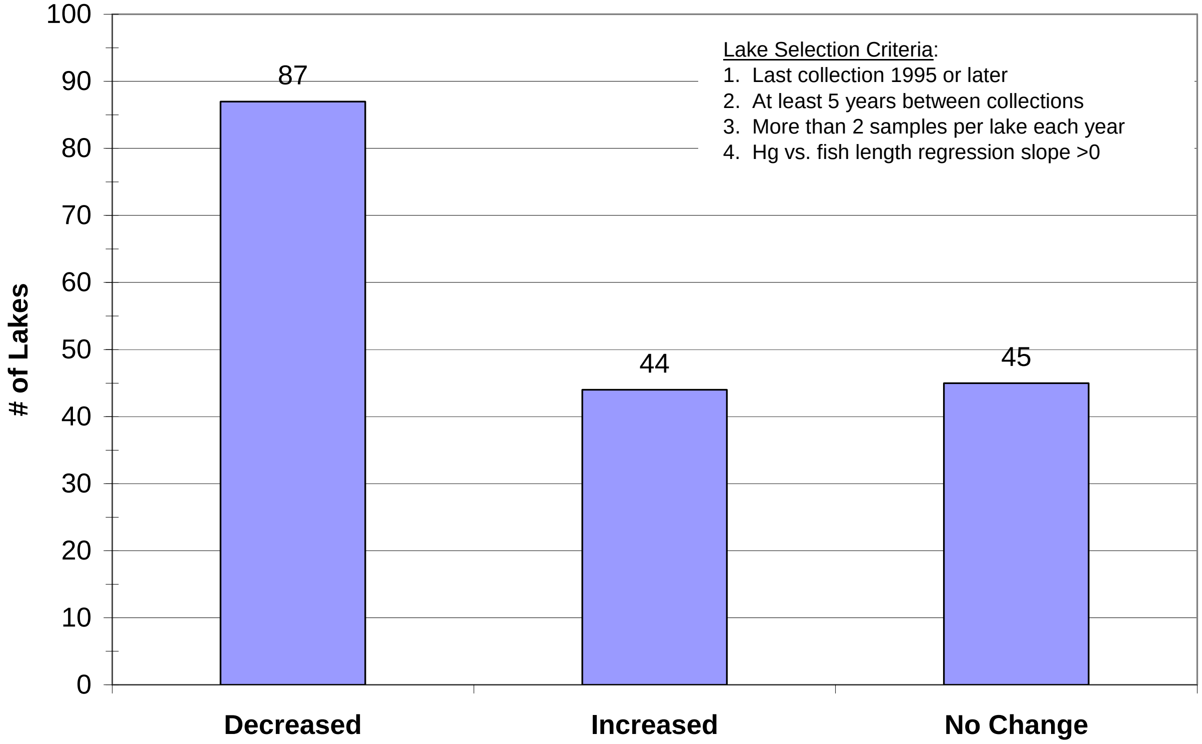


Figure 17 Comparison Between Recent and Historical Fish Mercury Levels in 176 lakes (Northern Pike and Walleye; Standard Lengths)

Selecting only the lakes with an historical collection around 1990 and a recent collection around 2000, the average change in fish mercury concentration was -11%; in other words, during the decade of the 1990s, there was a general decline in fish mercury concentrations of slightly more than one percent per year. Although it is not possible to relate the 68% reduction in state mercury emissions to this reduction in fish mercury levels during the same period because of the unknown response time, it does indicate there have been noticeable reductions in fish mercury levels that correspond with reductions in mercury emissions and deposition. The MPCA has a goal to continue that trend through the next decade (i.e., 10% reduction in fish tissue Hg concentration by 2010 compared to 2000).

6 TMDL Development

6.1 TMDL Formulation

The TMDL formulation used for these regional mercury TMDLs is similar to the approach used in the EPA-approved TMDL for the Ouachita River Basin, Arkansas (FTN 2002, FTN 2003). The Ouachita TMDL for lakes (FTN 2003) has a section, "TMDL Formulation," in which they define TMDL= (EL/RF) x SF, where EL is total existing load, RF is reduction factor, and SF is site specific factor. The Minnesota Mercury TMDL, Equation 6, uses the concept of total existing load (we call it "total source load") and reduction factor to define the needed TMDL. SF is not used in Equation 6, but the concept of a variable for site-specific factors was used previously (Section 5.2) with the introduction of runoff coefficient ("r") and bioavailability factor ("b"). As discussed previously, these factors cancel out of the equation that shows the change in fish tissue mercury is proportional to the change in deposition is proportion.

Annual loads are more appropriate than daily loads for mercury because the concern in this TMDL study is the long term accumulation of mercury rather than the short term acute toxicity events. The three-step process to determine the TMDL is (1) estimate existing load for point and nonpoint sources; (2) define the target loads; and (3) calculate load reduction factors necessary to achieve target values. The total source load and reduction factor are then combined to give the total maximum daily load in units of mass per time.

$$TMDL = TSL \cdot \left(1 - \frac{RF}{100}\right) \tag{6}$$

where, TMDL is total maximum daily load as an annual load (kg/yr); TSL is total source load during the baseline year; RF is the reduction factor, as a percentage. RF is based on the reductions needed to achieve target fish mercury concentrations (see Section 4.4). The RF accounts for differences in mercury transport and bioavailability of the estimated load for each region (see Section 5.2). Ultimately, the TMDL is presented in the basic equation form.

$$TMDL = WLA + LA + MOS \tag{7}$$

where, WLA is Wasteload Allocation (wastewater & stormwater sources), LA is Load Allocation (nonpoint sources), and MOS is Margin of Safety. Each of these TMDL components is discussed below.

6.2 Baseline Mercury Load for 1990

Total Source Load (TSL) of mercury, in kg/yr, is simply the sum of point source and nonpoint source loadings.

$$TSL = PSL + NPSL \tag{8}$$

where PSL is point source load and NPSL is nonpoint source load. PSL is estimated for each region based on the facilities total design flow and the average effluent mercury concentration.

$$PSL = \sum Q_i \cdot C_{avg} \tag{9}$$

where Q_i is the design flow for each permitted facility and C_{avg} is the estimated average mercury concentration in effluent (Table 7). Wastewater treatment plants (WWTPs) were separated into the two regions and their design flows were summed for each region for the wasteload calculations. A state average mercury concentration for wastewater effluent was used in the wasteload calculation, because the wastewater effluent was not considered different between regions. The design flow is a conservative assumption of WWTP mercury load because actual discharges are approximately 70% of design flows and very few WWTPs achieve their design flow. The current discharge information was used for the wastewater treatment plant design flow. Actual wastewater discharge has undoubtedly increased since 1990, with increasing population; however, the design flow is not considered an excessive overestimate because the number of wastewater treatment plants has not changed significantly since 1990 (see Section 5.5). A drinking water correction factor was applied to the point source load calculation to account for the one percent of influent to a wastewater treatment system that consists of drinking water. This one percent is not given as a credit because the 1990 total point source load was only 1.2% of the total mercury load.

Table 7 Mercury Load Inputs and Estimates for 1990

	SW			NE				
(a) WATER	Metro WWTP	Other WWTPs	Total	WLSSD WWTP	Other WWTPs	Taconite	Pulp & Paper	Total
No. Point Sources	1	515	516	1	98	19[3]	4	122
Mean Effluent Mercury Conc. (ng/L)	11	5		355	5	1.5	13	
Sum of Design Flows (mgd)	251	455	706	49	46	169	82	346
PS Mercury Load (kg/yr)[1][2]	3.8	3.2	7.0	23.7	0.7	0.3	1.5	26.2

(b) ATMOSPHERE	SW	NE
Atmospheric Deposition (g km^{-2} yr^{-1})	12.5	12.5
Region Area (km^2)	129,674	90,151
NPS Mercury Load (kg/yr)	1,621	1,127

(c) SUMMARY (kg/yr)	SW	NE
Point Source Load (PSL)	7.0	26.2
Nonpoint Source Load (NPSL)	1,621	1,127
Total Source Load (TSL)	1,628	1,153

[1] Effluent - drinking water correction factor is 0.99

[2] Anthropogenic sources to stormwater are included but the load is zero. Stormwater from precipitation is accounted for in the nonpoint source load.

[3] Seven taconite facilities each have 2-3 NPDES permits; 15 of the 19 facilities have design flows greater than 0.05 mgd.

Stormwater, if addressed in a TMDL, is usually included with the point source load and included in the wasteload allocation (see Section 5.5). Because the source of most mercury to stormwater is atmospheric deposition—the same as for nonpoint source loads—the atmospheric deposition source of mercury to stormwater is accounted for in the nonpoint source load calculation. The contribution of mercury from other sources to stormwater is accounted for in the wasteload calculation and is estimated to be insignificant (i.e., estimated as zero).

The only significant nonpoint source load (NPSL) is from atmospheric deposition:

$$NPSL = D_y \cdot SA_r \tag{10}$$

where D_y is atmospheric deposition (g km^{-2} yr^{-1}) and SA_r is regional surface area. The atmospheric mercury deposition for 1990 was 12.5 g km^{-2} yr^{-1}. The SW region covers 59% of the approximately 220,000 km^2 in Minnesota.

Based on these calculations of point source and nonpoint source mercury loads, the 1990 TSL was 1628 kg/yr for the SW region and 1153 kg/yr for the NE region. PSL was 0.4% of the SW region baseline load and 2.2% of the NE region baseline load. Combined, the water point sources contributed 1.2% of the baseline mercury load to the state's waters in 1990.

6.3 Wasteload Allocations (WLA)

The calculated allowable load (TMDL) of mercury that will not cause an exceedance of the applicable water quality standard is the sum of the load allocation (atmospheric deposition) and wasteload allocations (NPDES sources); therefore, the total allowable load must be apportioned between the atmospheric and wastewater point source loads. For both regions combined, the PSL was 1.2% of the total source mercury load. Clearly, all significant decreases in mercury loading will come from reductions in atmospheric deposition (i.e., load allocation). Therefore, the WLA is set at one percent of the TMDL or the 1990 PSL, whichever is lower. The resulting WLA is 4 kg/yr for the NE region and 7 kg/yr for the SW region. Assigning one percent to the WLA is equivalent to the cutoff USEPA's Mercury Maps model used to screen watersheds for significant point source impact (USEPA 2001d).

The WLA is by region and is not specific to each source, thereby providing a cap for the region that includes reserve capacity. Rather than assign an allocation to each source based on their current design capacity, continued mercury reduction will be encouraged through mercury minimization plans and enhanced phosphorus removal. EPA has determined, as a matter of policy, that NPDES point sources known to discharge mercury at levels above the amount present in the source water should reduce their loadings of mercury using appropriate, cost-effective, mercury minimization measures to ensure that the total aggregate point source mercury discharges are at a level equal to or less than the WLA specified in this TMDL. The reserve capacity in the WLA allows for permitting of additional wastewater discharges, but does not preclude the requirement of mercury minimization plans.

There are no known local hotspots. The MPCA looked at possible impacts from large water and air sources and did not see local impacts in elevated fish mercury concentrations compared to the background regional levels. The concern for local impacts from wastewater discharges are primarily where they discharge to effluent dominated streams (i.e., the dilution ratio is less than 10:1). In those cases, the NPDES permit requires stringent controls for solids and BOD removal. In the most restrictive cases the CBOD-5 limit is 5 mg/L. These restrictive effluent limits to protect receiving waters have the added benefit of low mercury concentrations in the effluent because most of the mercury is associated with solids. The MPCA has mercury data on two POTWs that have CBOD-5 limits of 5 mg/L. The mercury concentrations from these facilities are the two lowest values in a dataset of 30 POTWs: 0.90 ng/L and 1.30 ng/L.

There is an increasing use of the "Bio-P" process to meet 1 mg/L phosphorus effluent limits in Minnesota; at least 25 Minnesota facilities will be using it within their current permit cycle. This process greatly reduces solids and in so doing reduces mercury discharge. The MPCA has mercury concentrations from three facilities currently using Bio-P; the average mercury concentrations at the three facilities are 2.56 ng/L, 3.26 ng/L, and 3.65 ng/L. The MPCA will propose for rulemaking in 2006 that new or expanding dischargers get the 1 mg/L limit for total phosphorus if they discharge more than 1800 lbs TP per year.

In keeping with the adaptive watershed management approach and as part of the TMDL implementation plan, the MPCA will continue to investigate potential local impacts of point sources in effluent dominated streams, and where warranted, look to additional permit limitations and then, if necessary, develop a site-specific mercury TMDL.

6.4 Load Allocations (LA)

The total source load is the basis for the load allocation because all significant mercury load reductions are expected to come from atmospheric emission reductions. The regional reduction factors of 51% for the SW and 65% for the NE, multiplied by the total source loads yield mercury reductions of 830 kg/yr and 749 kg/yr, respectively (Table 8). Subtracting the load reduction goals from the baseline loads results in regional mercury load allocations of 798 kg/yr for the SW and 404 kg/yr for the NE. The needed reductions are similar for the two regions, but the mercury load allocation for the NE region is about one-half the SW region's load allocation.

Table 8 Mercury Load Reductions for Each Region

	Region	
	SW	NE
Total Source Load (kg/yr)	1628	1153
Reduction Factor (see Table 5)	51%	65%
Load Reduction Goal (kg/yr)	830	749
Mercury Load Allocation (kg/d)	2.18	1.10
Regional Area (km^2)	129,674	90,151
Mercury Deposition Goal (g km^{-2} yr^{-1})	6.1	4.4

Up to this point no assumptions are made about where the mercury reductions will come from, although it is clear from the estimate of total source load that the reductions must come from atmospheric deposition. If we therefore assume all mercury reductions must come from atmospheric deposition, dividing the mercury loading goal by the regional areas gives the areal mercury deposition goals of 4.4 g km^{-2} yr^{-1} for the NE region and 6.1 g km^{-2} yr^{-1} for the SW region.

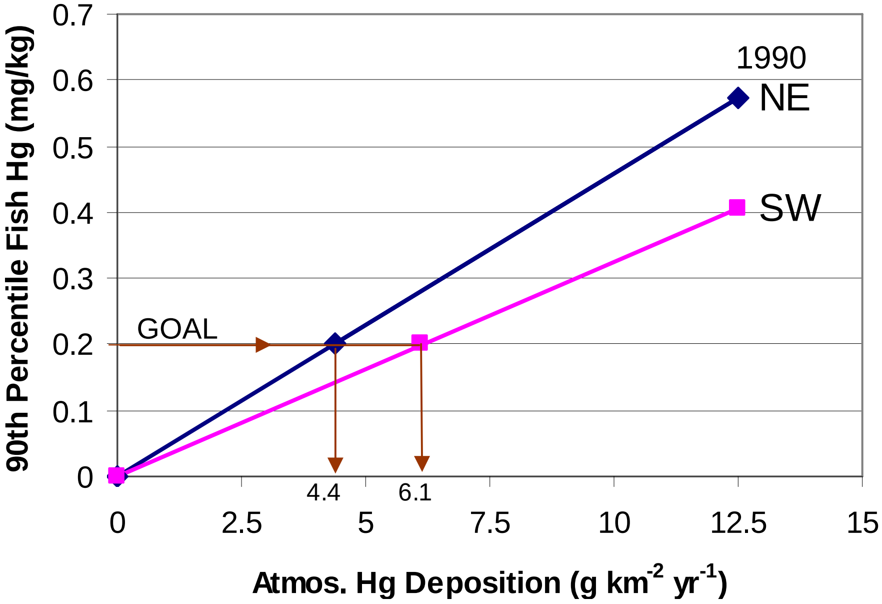


Figure 18 Proportional relationship between mercury deposition and fish mercury concentration in the two TMDL regions

The reductions in fish mercury concentrations (90th percentile) in proportion to atmospheric mercury deposition are illustrated for each region in Figure 18. Atmospheric mercury deposition in 1990 was 12.5 g km^{-2} yr^{-1} for both the NE and SW regions; while the 90th percentile mercury concentrations for standard size walleye were 0.572 mg/kg in the NE region and 0.405 mg/kg in the SW region. As discussed in Section 5.2, mercury concentrations in the fish are expected to eventually decrease in proportion to the decrease in atmospheric mercury deposition. The lines connecting the 1990 fish mercury concentrations to the graph's origin represent the proportional decrease. The ultimate goal is to achieve 0.2 mg/kg fish mercury level in both the NE and SW, which requires a 65% decrease in the NE and a 51% decrease in the SW. Assuming the mercury deposition continues to be uniform across the state, the NE will not meet its goal when the SW has met its goal; therefore, the lower mercury deposition goal of 4.4 g km^{-2} yr^{-1} becomes the necessary goal for the whole state.

As described in Section 5, atmospheric sources of mercury in Minnesota are categorized as 70% anthropogenic and 30% natural. Natural sources cannot be controlled and are expected to remain at the same long term average; therefore, all mercury reductions must come from anthropogenic sources. Taking load reductions from anthropogenic sources results in a 93% reduction for the NE and 73% reduction for the SW (Table 9). State sources are 10% of all mercury deposition (0.25 • 40% regional sources) and 14% of the anthropogenic sources (10% ÷ 70% = 0.143).

Table 9 Mercury Load Allocation for In-State and Out-of-State Emissions

	TMDL Region	
	SW	NE
Load reduction from anthropogenic sources (RF ÷ 0.7)	73%	93%
State's fraction of anthropogenic sources (10% ÷ 70%)	0.143	0.143
In-State contribution to the TMDL Load Allocation (0.143 • LA)	0.31 kg/d (249 lb/y)	0.16 kg/d (126 lb/y)
Out-of-State contribution to the TMDL Load Allocation [(1- 0.143) • LA]	1.86 kg/d (1,495 lb/y)	0.94 kg/d (756 lb/y)

Consequently, the in-state load allocations of the TMDL are 0.31 kg/d and 0.16 kg/d for the SW and NE regions. The remaining load reductions must come from outside the state and are, therefore, the out-of-state load allocations.

The state's load reduction goals can be translated to emission reduction goals based on the 1990 baseline year emissions and deposition. Atmospheric deposition of mercury is considered uniform across the state, and emissions dispersion within the state overlaps the two regions; therefore, the greater regional reduction goal must be applied to a statewide emissions goal to achieve the goal in the more sensitive region.[3] Total mercury emissions in Minnesota were estimated to be 5113 kg in 1990. A 65% reduction in total deposition translates to a 93% reduction of anthropogenic deposition. The state's contribution to emissions must be reduced 93%, along with other anthropogenic sources to meet the 65% reduction in total deposition (Table 10). The state's emissions must be reduced from 11,272 lb (5113 kg) in 1990 to 789 lb (358 kg) as a final annual emissions goal (Table 10).

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[3] This approach is consistent with the accepted approach to environmental protection; for example, dissolved oxygen (DO) water quality standards are applied at a point in a receiving water where the DO concentration is lowest (the "DO Sag") or to the early morning hours when DO is lowest within a diurnal cycle.

Table 10 Minnesota's Mercury TMDL Emissions Reduction Goal

	Annual Statewide Mercury Emissions	
State mercury emissions for 1990	11,272 lb	(5,113 kg)
Mercury Emissions <u>Reduction Goal</u> (0.93 • 1990 emissions)	10,483 lb	(4,755 kg)
Mercury <u>Emissions Goal</u> (1990 Emissions – Reduction Goal)	789 lb	(358 kg)
Emissions reduction as of 2005 (70% of 1990 emissions)	7,931 lb	(3,597 kg)
Emissions reduction remaining as of 2005 to achieve goal	2,552 lb	(1,158 kg)
Percent of 1990 Emissions Reduction Goal remaining as of 2005	24%	

Based on the mercury emissions inventory for 2005, 76% of this emissions reduction goal has been achieved; therefore, as of 2005, 24% of the reduction goal remains.

6.5 Reserve Capacity

Reserve capacity refers to load that is available for future growth when actual loads are less than the load allocation. There is no reserve capacity for nonpoint sources, because actual nonpoint source loads are far in excess of the Load Allocation. Reserve capacity is available for water point sources, because the actual mercury load is less than the Wasteload Allocation. The wasteload allocation for this mercury TMDL is set at one percent of the TMDL or equal to the 1990 point source load, whichever is lower. The one percent level is 4 kg for the NE region and 8 kg for the SW. The 1990 point source load in the SW was 7 kg; therefore, the wasteload allocation is set at 7 kg rather than 8 kg. One percent is considered a *de minimus* level, meaning mercury load at, or below, this level is insignificant. Therefore, any changes in load below the *de minimus* level are insignificant. The estimated mercury point source load in 2004 was about 7 kg for the state, resulting is a reserve capacity of about 4 kg. The mercury TMDL implementation plan will establish a procedure to account for the reserve capacity potentially available for new and expanded sources.

7 Margin of Safety

A Margin of Safety (MOS) is required in the TMDL to account for uncertainty in the TMDL calculations. MOS can be either explicit (e.g., additional 10% load reduction), implicit in the calculations, or a mix of the two. In conventional TMDLs, uncertainty arises through the use of water quality models and Best Management Practice (BMP) choices. An implicit MOS can include using the most conservative parameters when there is a range from which to choose or using the most conservative effectiveness assumption when selecting BMPs. In this mercury TMDL, uncertainty arises from the mercury movement through the environment, and, in particular, the conversion of inorganic mercury to methylmercury. Very important, but still poorly understood, are the sulfate-reducing bacteria, which methylate the mercury and are abundant in wetlands.

The greater abundance of wetlands in the NE region and the associated sulfate-reducing bacteria is thought to be one of the major reasons why fish mercury concentrations are significantly higher than in the SW region. Research by the Agency and others are actively pursuing this important link between sulfate reduction and fish tissue methylmercury levels. Lowering sulfate load to wetlands could reduce the sulfate-reducing bacteria activity and thereby reduce the methylation of inorganic mercury. Consequently, the needed mercury load reduction would have to be less.

We know sources of atmospheric sulfate are actively being reduced through a variety of activities at the federal and state levels. We believe the reduction in fish mercury through reductions in sulfate could be significant. The Agency believes the relationship between sulfate concentrations and methylmercury concentrations is adequately established, but not to the degree that we can determine an explicit MOS. Thus, for this initial phase of the TMDL, it will be the primary implicit MOS for the NE region.

The SW region has an explicit MOS, because the TMDL goal is a statewide 93% reduction in mercury emissions. This is protective of the NE region but over-protects for the SW region, which has only a 73% mercury emission reduction goal. Uniform deposition across the entire state is the reason for the more protective requirement in the SW region.

8 Seasonal Variation and Critical Conditions

Seasonal variations and "… critical conditions for stream flow, loading, and water quality parameters" are discussed in 40 CFR 130.7(c)(1). Fish accumulate enough mercury over the years of their lifespan to become a health hazard to humans and wildlife. Mercury deposition and water concentrations fluctuate based on seasonal rainfall patterns; however, seasonal variations are not significant to this TMDL because it is expressed as an average annual load. The mercury concentration in the fish represents an integration of all temporal variation up to the time of sample collection. Variability among fish because of differences in size, diet, habitat, and other undefined factors are expected to be greater in sum than seasonal variability.

Critical conditions, such as dawn period for low dissolved oxygen, are not relevant to mercury in fish because the fish tissue concentrations reflect integration over time of various factors. However, there are critical conditions in the sense of water bodies that are more sensitive to mercury loading because of their water chemistry. This aspect of critical conditions has been addressed in this TMDL by using the regional approach and acknowledging that the NE region is more sensitive to mercury loading than the SW region.

9 Final TMDL

The conventional equation for a TMDL is as follows: TMDL = WLA + LA + MOS. As described above, the Margin of Safety (MOS) is implicit for the NE region and, therefore, not quantifiable. The MOS for the SW region was given in terms of the anthropogenic emissions reduction goal (i.e., 93% reduction matching the NE, rather than 73%). If the MOS for the SW were interpreted as applying the NE Reduction Factor of 65%, instead of 51%, the LA for the SW is reduced from 2.16 kg/d to 1.55 kg/d, which leaves a MOS of 0.62 kg/d. The NE and SW regional TMDLs fit the TMDL equation as follows:

$$\text{NE Region TMDL (1.10 kg/d)} = \text{WLA (0.01 kg/d)} + \text{LA (1.09 kg/d)} + \text{MOS (implicit)} \qquad (11)$$

$$\text{SW Region TMDL (2.18 kg/d)} = \text{WLA (0.02 kg/d)} + \text{LA (1.55 kg/d)} + \text{MOS (0.61 kg/d)} \qquad (12)$$

The WLA is defined for these mercury TMDLs as one percent of the TMDL to ensure that water point source mercury load remains *de minimus*.

As required by Federal law, this TMDL is not applicable to waters in Indian Country.

10 Monitoring & Research Plan

Monitoring to detect environmental change to changing atmospheric mercury deposition will follow the recommendations of Mason *et al.* (2004). Monitoring options that are being considered include the following:

- Fish contaminant monitoring of previously sampled lakes and rivers (this is ongoing)
- Sentinel lakes: 4-5 lakes around each of the MDN sites in Minnesota; monitor air, water, & fish tissue (biopsy)
- Lake sediment cores and recalculation of mercury deposition for representative lakes
- NPDES upstream/downstream monitoring for traditional wasteload allocation studies
- Continued air monitoring for wet deposition; new monitoring stations required for dry deposition and urban areas

The MPCA and its research partners in Minnesota are studying factors affecting mercury contamination of fish. Widely cited Minnesota research in the 1990s analyzed lake sediment cores to estimate historical mercury deposition and its sources. Current work is focused on understanding the local factors, such as land cover effects and food chain structure, which might explain the observed variability in mercury bioaccumulation among lakes. Another research project in the state is testing the effect of increased sulfate deposition on mercury methylation in a wetland (Jeremiason *et al.* 2003). The outcomes of these studies will help refine the implementation of the mercury TMDLs.

11 Projected Implementation

11.1 Background

According to USEPA's *Guidance for Water Quality-based Decisions: The TMDL Process* (EPA 440/4-91-001), TMDL development activities consist of the following process – selection of the pollutant to consider, estimation of the waterbody's assimilative capacity, estimation of the pollution from all sources to the waterbody, determination of total allowable load, and, finally, allocation (with a margin of safety) of pollution reductions such that water quality standards are met. This has been accomplished in the preceding sections.

Implementation planning is not a formal requirement of the TMDL process. This is very clear in Figure 2 on page 21 of the 1991 TMDL Guidance document noted above – detailed implementation planning occurs after the TMDL plan is approved by USEPA. The Clean Water Act describes implementation planning in the section following TMDL requirements, Section 303(e). However, it is has long been recognized that the TMDL planning document should contain at least the outline of the specific implementation planning needed to meet the pollution reduction goals set in the TMDL. This informs the public and aids in the discussion on reasonable assurance.

Once this TMDL pollution reduction plan is approved by USEPA, the reduction goals are legally enforceable to the extent that legal authorities exist. This TMDL will not generate new legal authorities. Draft implementation planning is not legally enforceable – they are approaches that the Agency believes are appropriate, but subject to change based on further refinement as the implementation planning becomes more developed.

11.2 General Approach

The best available information was used to establish this TMDL; nevertheless, uncertainty warrants an adaptive watershed management approach to implementation (Dilks and Freedman 2004). The TMDL implementation will incorporate the adaptive watershed management approach by establishing a monitoring plan, interim targets, and a timeline. Future information and analysis may warrant revisions in the goals or the tools used to assess progress toward the goals.

An important component in developing a strategy to meet water-quality standards is to consider the value and effectiveness of efforts already in place. As noted previously, from 1990 to the present Minnesota's mercury emissions were reduced more than 70%. This section summarizes initiatives that the MPCA believes have already reduced fish contamination in Minnesota and will maintain a path of reduced fish contamination in the future. In addition to reductions from Minnesota sources, reductions in air emissions of mercury by national and international sources will be needed to meet Clean Water Act standards in Minnesota, because there is significant mercury deposition in Minnesota from these sources.

Reduced deposition of mercury is projected to have beneficial human-health and environmental effects, which produce economic benefits. To estimate the total economic value of improvements in environmental goods and services, a state-wide contingent-valuation study was completed in 1999 (Hagen *et al.* 1999). The study was designed to elicit the willingness of Minnesota households to pay for reductions in adverse health and environmental effects that result from mercury deposition. A major conclusion was a best estimate annual state willingness to pay of $212 million for a policy that was projected to reduce mercury deposition in Minnesota by approximately 12%. The willingness to pay was greater for greater reductions in mercury deposition; therefore, it follows that the reductions outlined in this TMDL report (i.e., 65% reduction in mercury deposition) would translate to a much greater economic benefit within the state. These reductions will also significantly reduce health risks associated with ingesting fish containing excess mercury.

11.3 Implementation for Wasteload Allocation

Despite the relatively very minor contribution of point sources to the total mercury load, the Agency will continue to strive to reduce mercury from all sources inside the state, including point source water discharges. To do so, the permitted facilities will establish mercury minimization plans (Table 11). In receiving water segments that are discharge-dominated, the dischargers will be subject to permit conditions that address ambient fish tissue monitoring. This monitoring will determine the relationship of the *de minimus* assumption with actual fish tissue conditions on a more localized basis and could result in mercury limitations in NPDES permits.

Table 11 WWTP Mercury Reduction Plan

WWTP Average Wet Weather Flow (g/d)	Sampling per Year	Mercury Minimization Plan (MMP) Required
Less than 200,000	0	NO
200,000 – 500,000	1	YES
Greater than 500,000	4	YES

The sum of municipal WWTP, stormwater & industrial WWTP WLA loads shall remain *de minimus*, i.e. less than 1% of the atmospheric deposition load or 11 kg per year. This includes both existing WLA facilities, expanding facilities, and new facilities. MMPs and monitoring will be required for those facilities with a wet weather design flow of 0.2 mgd or greater. MMPs will include minimization of mercury in solids as well as in discharge.

Additional restrictions will be required if the *de minimus* is exceeded. For new/growth permits in the SW region, the focus will be trading, either with [1] NPS sediment reductions (the ratio must be agreed upon), [2] stormwater sediment reductions in the areas that are growing (the ratio must be agreed upon), and/or [3] trade-up in treatment to Bio-P or Chem-P, either of which will reduce the effluent mercury to below the new WQS after out-of-state reduction goals are met. Bio-P seems to reduce mercury to about one-half to one-third of the previous average effluent concentration.

For the NE region, new/growth permits will focus on trading, either with [1] air sources for sulfur reductions and/or mercury reductions, [2] stormwater sediment reductions, and/or [3] inflow/infiltration (I/I) reductions. Bio-P is also an important option.

The exception to the *de minimus* provisions for the WLA portion of this TMDL originates with the water quality rules for the Lake Superior Basin, Chapter 7052, and the GLI Guidance from which it was derived. Provisions for the phase-out of mixing zones for existing dischargers by March 23, 2007, and for not allowing mixing zones for new or expanded dischargers at commencement of discharge are specific to GLI and Chapter 7052. These provisions apply when establishing a TMDL. Therefore, all dischargers in the Basin will initially or eventually need to meet the 1.3 ng/l mercury water quality standard and mass caps as a waste load allocation for their discharge.

11.4 Implementation for Load Allocation

Minnesota clearly is dependent upon other areas to reduce emissions, and Minnesota has set an example that other areas should emulate. Minnesota has, through both voluntary and regulatory approaches, reduced mercury emissions by 68% between 1990 and 2000 (See section 5.3.1), and the MPCA expects to meet the mandated reduction goal of 70% by 2005. (The 68% reduction of the 1990 mercury emissions is equal to 73% of the mercury TMDL reduction goal.) Mercury reductions outside the State have not been quantified, although USEPA reports national emissions have been reduced at least 45% between 1990 and 1999 (See section 5.3.1).

Attempting to reduce Minnesota's fish contamination by exporting more of our emissions—through higher stacks or intentional conversion of divalent mercury emissions to elemental mercury—invites other areas to also export mercury, which probably would result in greater fish contamination in Minnesota. Therefore, Minnesota's mercury TMDL emission reduction strategy is to calculate the percent reduction that is needed and reduce mercury emissions by that degree without regard for where the emissions will be deposited.

To maintain economic fairness while focusing on meeting water quality standards, the Agency will employ a phased approach to achieve a goal of 789 pounds of mercury emitted from all Minnesota sources. Short-term actions, check-in points, and future action options are described below. The sector-specific reduction milestones will be used as guideposts by the MPCA to decide whether reductions are on track in the future. If emission reductions are not on track, the MPCA will review the current state of knowledge about mercury deposition and control options and costs, and develop a strategy to obtain further reductions to achieve the milestone. The strategy will contain regulatory controls as necessary to reach target goals.

Recognizing the overall goal of reducing mercury deposition in the state, one sector might reduce more than the sector goal, while another sector is not able to reduce as much, yet the reduction strategy may still achieve its goal. The same is true for the ratio of in-state to out-of-state reductions. If national, international, and other state reductions are large enough, those reductions may obviate the need for some reductions in Minnesota. This is important because over the past decade Minnesota has reduced mercury emissions using cost-effective steps that others may not yet have taken.

Section 12 outlines the mercury reduction programs underway in Minnesota, nationally, and internationally. It provides the assurance that several robust mercury control programs are well-established and have been very effective. Section 5 provides context that fish tissue concentrations have been in decline in recent years, mercury emissions are trending down in all relevant emission inventories, and that reduced sulfate concentrations from sulfur dioxide emission reduction programs may magnify the benefit of reduced mercury emissions.

After finalizing and submitting this TMDL to USEPA for approval, the MPCA will begin to work with stakeholders to develop an implementation plan. As explained above, the implementation plan is not a formal requirement of the TMDL process. The TMDL is expected to contain an outline of the specific implementation planning needed to meet the TMDL goal. The MPCA expects to conduct in-depth implementation planning in 2005-2006 after this TMDL is approved. The implementation planning process will need to involve a careful review of the cost and efficacy of technically available control options, and the development of federal regulations that would reduce mercury emissions from source categories like power plants, mini-mills and industrial boilers.

11.4.1 Short-term Actions

New and Expanding Sources of Mercury to the Atmosphere

To limit growth of mercury emissions because of construction of **new** or **expanding** emission sources in Minnesota, the MPCA will develop a permitting strategy for new and/or expanding air emissions sources of mercury that considers the following:

- Establishing an appropriate facility *de minimus* emissions rate
- Requiring new or expanding sources to use state-of-the-art mercury control technology if the *de minimus* rate is not feasible/achievable/possible
- Investigating how to allow offsetting reductions

Develop Monitoring and Reporting Protocol

The MPCA will work with air emission sources to develop a monitoring and reporting protocol with the goal of better quantifying annual mercury emissions. An improved Minnesota air emissions inventory is needed to track progress at meeting reduction targets.

Current Reduction Strategies

The MPCA will continue to employ strategies already in place as described below to continue the trend of decreased air emissions. The MPCA expects interim and final targets will be met, especially considering pending federal regulations and a strong commitment to voluntary reductions by existing facilities.

Continue existing state regulations and programs to collect mercury. Existing regulations, including product bans, disposal requirements and pollution control equipment at waste combustors will continue to reduce product-sector emissions. Additionally, the MPCA and the Minnesota Office of Environmental Assistance continues to identify and provide education to industrial sectors and Minnesota citizens about proper storage and management of mercury-bearing wastes.

Support voluntary reductions. Under the MPCA’s current mercury strategy, the MPCA expects facilities to voluntarily reduce air emissions whenever possible. The MPCA will continue to work with air emission sources to encourage reductions.

Encourage the development of federal regulations: MPCA believes that a national program regulating mercury emissions from existing and future emission sources holds the most promise, by addressing the substantial contribution by sources outside of Minnesota to Minnesota water bodies while minimizing competitive disadvantages that a state-level only regulation could create. The MPCA will continue to work with EPA to implement control standards/programs that require substantial reductions from electric utilities, steel mini-mills, crematories and other emissions sources across the country. The MPCA filed detailed comments with USEPA in 2004 urging promulgation of strong federal mercury reduction requirements for the electric power sector.

Evaluate impacts of federal regulations. The MPCA will continue to assess the impacts of federal actions to inform potential additional state regulatory actions. In particular, the MPCA will closely follow the development and implementation of the federal mercury emission reduction program for power plants: the Clean Air Mercury Rule. The Implementation Plan will thus be able to consider the effect of that standard on power plant emissions. At the same time, USEPA is finalizing a new regulation that would reduce sulfur dioxide emissions in the eastern US by 70%. MPCA will continue to investigate whether sulfur dioxide reductions of this scale will magnify the effect of mercury emission reductions, and the impact of the program on Minnesota's reduction targets.

Investigate regional state cooperation. In the absence of strong federal regulations, reductions in Minnesota may be more effectively and equitably achieved by cooperating with other states in the region to develop strategies to reduce emissions sources. The MPCA will actively coordinate the above strategies and consider regional regulations if needed to meet reduction targets.

11.4.2 Reduction Targets and Milestones

Substantial reductions are necessary from existing sources to achieve a 93% reduction in overall anthropogenic emissions from 1990 levels. To initiate development of an implementation plan, the MPCA has set overall and sector-specific reduction targets (Table 12) for the in-state energy, taconite and products sectors with interim outcome-based targets. These targets will guide the MPCA in its implementation planning. They will aid in establishing set intervals for the MPCA to evaluate progress, reviewing recent developments in controlling mercury emissions, and assessing on new strategies.

An example of ways that future federal regulations might affect Minnesota's goals is the proposed mercury cap and trade program. Mercury emission control on power plants is expensive, and becomes more expensive as the reduction target moves from 70% to 80% to 90%. The Acid Rain program of the 1990 Clean Air Act Amendments showed that a cap and trade program is extremely cost-effective in meeting emission reduction targets because it drives reductions to the place where larger, cheaper reductions can be made. The USEPA has adopted a cap and trade program as the basis for the Clean Air Mercury Rule (issued March 15, 2005). In the Mercury TMDL implementation plan, the MPCA will consider the appropriateness of keeping a state sector target if the national trading program will assure that the national power sector will reach an adequate percentage reduction in mercury emissions.

Table 12 Summary of Reduction Targets

Sector	1990 Emissions	2000 Emissions	Target #1	Target #2	Target #3
Energy	1,667	1,834	675	470	313
Material Processing	723	758	550	280	138
Products	8,881	1,045	475	350	338
All sources	11,272	3,638	1,700	1,100	789

For Target #1, when the national emission reductions are expected to reach 65% from 1990, the state target of 1700 pounds [Target #1] must be reached. If the state target is not met, regulatory tools will be developed as necessary to reach target goals, unless achieved national reductions exceed their target and obviate the need for some of the state reductions.

For Target #2, when the national emission reductions are expected to reach 80% from 1990, the state target of 1100 pounds [Target #2] must be reached. If the state target is not met, regulatory tools will be developed as necessary to reach target goals, unless achieved national reductions exceed their target and obviate the need for some of the state reductions.

For Target #3, when the national emission reductions are expected to reach 93% from 1990, the state target of 789 pounds [Target #3] must be reached. If the state target is not met, regulatory tools will be developed as necessary to reach target goals, unless achieved national reductions exceed their target and obviate the need for some of the state reductions.

11.4.3 Regulatory Actions

The MPCA has identified potential regulatory options for ensuring reasonable progress towards achieving the targets of the TMDL, some of which were selected from the 1999 Mercury Contamination Reduction Initiative Advisory Council's *Source Reduction Feasibility and Reduction Strategies (SRFRS) Committee Report on Options & Strategies for Reducing Mercury Releases*[4]. If current reduction strategies, including upcoming federal regulations and voluntary measures, under-achieve by not reaching interim or final target goals, the agency will need to consider further voluntary strategies and enact regulatory approaches to meet TMDL reduction requirements to ensure that the final reduction target are reached. These strategies might include:

- Technology-based limits for emissions sources
- Establish a sector-level cap
- Establish a state-level cap and trade system
- Impose fees on mercury emissions

12 Reasonable Assurance

12.1 Assurances Occur at Multiple Levels

A complete TMDL evaluation requires reasonable assurance that the impaired waters will attain water quality standards. When water point sources dominate as pollutant sources, reasonable assurance is straightforward: implement reduction in NPDES permits. This is not the case when nonpoint sources are the major source of the pollutant. For this Minnesota Mercury TMDL, there are reasonable assurances in the past actions on state, federal, and international levels that had a measurable affect on sources of mercury. Can we also see a measurable affect on environmental concentrations of mercury, and especially on the endpoint: mercury in fish? The first subsection below summarizes what reasonable assurances we can determine from research and monitoring of mercury in the environment. Other aspects of reasonable assurance are present and future decisions: will actions to reduce mercury contamination continue, and what new or proposed actions will reduce mercury? Subsections below summarize what we consider positive steps to reduce mercury on the three governmental levels.

12.2 State Level Assurances

Minnesota has been a national leader in reducing mercury releases to the air, water and land since the early 1990s. The state employs an array of voluntary, regulatory, incentive-based and educational tools that involve local governments, state agencies and businesses. In concert with similar initiatives on the Federal level, Minnesota's efforts have contributed to a 70% reduction in mercury emissions over the last 13 years (Table 13). The Minnesota Mercury Emissions Inventory (MPCA 2005) describes the specific statewide mercury emissions in 1990, 1995, and 2000. The MPCA mercury emission inventory has an earlier baseline and is more inclusive than the USEPA inventory. The USEPA inventory tends to exclude sources that are more difficult to quantify or verify. The best example of such a source is the large amounts of mercury that must have volatilized from latex paint until it was no longer added to paint after 1992.

Carol Hubbard and Clancy inspecting a classroom for mercury

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[4] MPCA, 1999. available at http://www.pca.state.mn.us/air/mercury-mn.html#initiative

Table 13 Summary of Mercury Reduction Strategies Used in Minnesota Since 1990

Voluntary Programs	
Health Care Outreach	Education on management & reduction of mercury-containing equipment.
Household/Small Business Hazardous Waste Collection	Many county-run programs accept mercury-containing items from homeowners and businesses.
Dental Office Outreach	Municipal wastewater treatment plants and the Minn. Dental Assoc. established best management practices and goals for 100% participation.
Thermostat Take-back	Through a reverse distribution system involving contractors and wholesalers, manufacturers take back out-of-service units.
Mercury Switches in Automobiles	Law requires “good faith effort” to remove mercury switches prior to crushing; bounty of $1/switch offered by major steel recycler.
Mercury-Free Zone Program	Schools pledge to be mercury free and receive an assessment and educational visit by the MPCA’s mercury educator, Carol Hubbard, and Clancy, its mercury-detecting dog.
Voluntary Reduction Agreements	Large emitters enter into voluntary agreements to reduce emissions. http://www.pca.state.mn.us/air/mercury-mn.html
Regulatory Programs	
Waste Combustor Standards	Sets air emission limits on mercury and requires mercury reduction plans for municipal and medical waste incinerators.
Water Discharge Standards	Waste water dischargers are required to monitor for mercury using EPA Method 1631; mercury effluent limits are set in some cases
State Laws	
Fluorescent Lamp, Other Product Disposal Ban	Requires businesses and households to recycle fluorescent lamps, stimulating development of recycling infrastructure.
Mercury-containing Product Bans	Toys, games, apparel and thermometers that contain mercury may not be sold in Minnesota
Dairy Manometer Ban and Buy-back	Bans the sale, installation and repair of mercury-containing manometers, establishes $100 incentive for turning in old gauge.
Relay Manufacturer Responsibility	Requires manufactures of mercury displacement relays to provide education and incentives, cover costs of managing out-of-service units.
Battery Mercury Reduction	Bans mercuric oxide batteries and the addition of Hg to alkaline batteries. Establishes a 25-mg limit in button batteries.
Mercury in Construction/Demolition	Law prohibits disposal, implying removal prior to demolition. Education and enforcement conducted.
Mercury Reduction Law (1999)	Requires the state to pursue Advisory-Council-recommended strategies, establishes a goal of 70% reduction in emissions by 2005 based on 1990 levels. Final report due in 2005.

Many of Minnesota’s mercury-reduction initiatives have become national models. For example, the thermostat industry’s voluntary take-back program for mercury thermostats started as a collaborative pilot in Minnesota and is now a nationwide program. The Minnesota Legislature passed some of the first laws in the country banning mercury in certain products, some of which (e.g., zero mercury in alkaline batteries) led to similar national laws.

Minnesota also adopted rules setting standards for municipal and medical waste incinerators ahead of federal requirements that call for stricter emissions limits than the current federal standard. These waste combustor standards, coupled with increased mercury product management and reduction, led to a 93%

decrease in emissions from municipal and medical waste incinerators, dropping from 1,053 kg (2,322 lb) in 1990 to 76 kg (167 lb) in 2000.[5]

In 1996, the MPCA initiated the Mercury Contamination Reduction Initiative aimed at reducing mercury contamination of fish in Minnesota lakes and rivers. As part of the initiative, the agency formed a stakeholder advisory council to develop recommendations on mercury-reduction strategies. The advisory council's recommendations were adopted by the Minnesota Legislature in 1999 and continue to form the basis of Minnesota's mercury-reduction program. These strategies include establishing reduction goals, national and international strategies, research, reducing purposeful use, and voluntary agreements. Minnesota's statewide mercury-reduction goal, recommended by the advisory council and set in state statute, is to reduce annual mercury releases 60% by 2000 and 70% by 2005, compared to 1990 levels. The MPCA estimates statewide emissions in 2000 of 1,650 kg (3,638 lb), a 68% reduction from estimated 1990 levels. Minnesota mercury emissions sources and trends are discussed in more detail in Section 5.3.1.

The 1999 law established a voluntary mercury-reduction agreement program encouraging the largest emitters in the state to enter into agreements with the MPCA to voluntarily reduce their mercury air emissions. Participants in the program are expected to implement cost-effective, technologically feasible reduction measures. The MPCA agreed not to pursue additional state regulations, at least until 2005, as long as adequate progress is made in reducing emissions (MPCA 2002).

Twelve companies and two regional waste management jurisdictions participate in the voluntary agreement program and have implemented steps to reduce emissions, pledged reductions or engaged in research in the hope of discovering future reduction strategies. Progress reports are available at http://www.pca.state.mn.us/air/mercury-mn.html. When fully implemented, reduction agreement actions initiated to date will result in additional reduction in annual emissions of an estimated 155 kg (342 lb) by 2008, or about 9.4% of 2000 emissions. These actions include fuel switching and increased controls by electric utilities [a 120 kg (264 lb) annual decrease, or 16% reduction for the sector] and added controls on sewage-sludge incineration [35 kg (78 lb) or 70% sector reduction]. In addition, participants have removed from their plants and properly managed hundreds of pounds of mercury-containing equipment, mercury that could have been released to the environment.

Minnesota continues to work with stakeholders to reduce mercury releases to the environment from product-related uses, taconite processing and energy production. Continued reductions in product-related emissions are expected as mercury use in products declines and sound management increases. However, achieving significant reductions beyond the progress to date will require reductions in mercury emissions in the taconite and power-generation sectors. Currently, no cost-effective reduction technologies have been identified for taconite-processing operations. The MPCA and the Minnesota Department of Natural Resources are working with Minnesota's taconite industry to identify future control technologies and funding research to develop these technologies.

In 2000, as part of a commitment to reduce emissions under the state's voluntary mercury-reduction agreements, Minnesota Power substituted lower-mercury coal to achieve a 32 kg (70 lb) annual reduction in mercury emissions from its operations.

In December 2003, the Public Utilities Commission approved Xcel Energy's Metropolitan Emissions Reduction Program (MERP) that will re-fire two coal plants with natural gas and upgrade the pollution-control equipment at a third, metro-area plant. When fully implemented in 2009, the MERP will result in an estimated annual mercury reduction of 77 kg (170 lb).

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[5] Changes in mercury emissions after 1990 are not included in the TMDL calculation, since the TMDL is based on the 1990 baseline year.

Taken together, Minnesota Power's lower-mercury coal and Xcels' MERP will result in a reduction of 109 kg (240 lb), a 16% reduction in utility-sector emissions and a 6.3% reduction in total emissions compared to 2000 levels. These reductions will account for a 2% reduction in total emissions when fully implemented (based on 1990 levels). The MPCA is awaiting the outcome of Clean Air Act standards development for power plants and several multi-pollutant proposals in Congress that have the potential to significantly decrease mercury emissions from power generation.

Metropolitan Council Environmental Services (MCES) operates two sewage sludge incinerators. Mercury emissions from these plants dropped from 112 kg (247 lb) in 1990 to 51 kg (112 lb) by 2000 largely because of reducing mercury inputs to the wastewater. The MCES is constructing a new sewage sludge incinerator at its metro plant (scheduled to go on line in mid-2004) and expects mercury emissions to be reduced by approximately 35 kg (78 lb).

MPCA will propose for rulemaking that new or expanding dischargers get 1 mg/L limit for total phosphorus if they discharge more than 1800 lbs phosphorus per year. The new limit will result in more facilities adding Bio-P or Chem-P processes to reduce phosphorus; these processes have already been added to a number of large WWTP in Minnesota and data collection has shown reductions in mercury concentrations as well, by one-half to two-thirds. Therefore, continued mercury reductions from WWTPs are expected as the 1 mg/L phosphorus limit is implemented.

12.3 National and International Assurances

Because of long-range transport of mercury in the atmosphere, reductions in mercury air emissions outside of Minnesota will eventually lead to reduced mercury deposition in Minnesota and reduced contamination of Minnesota fish. A variety of programs, initiatives and regulations exist in North America and internationally to reduce mercury emissions (Table 14). Notably in 2003, the United Nations Environmental Programme determined that mercury is a Pollutant of Concern that warrants international action and established a mercury program to assist developing countries with identification and reduction of mercury emissions. Given these programs, the success of mercury-reduction initiatives in developed countries and the eventual transfer of technology to the developing world, the MPCA expects that these initiatives will eventually lead to reductions sufficient to reduce fish mercury concentrations in Minnesota water bodies. However, uncertainties about the timing of implementation of reduction efforts world-wide and the complexity of mercury cycling make it difficult to predict when the effects of these actions will result in significant improvements in Minnesota.

Minnesota participates, where it can, in these national and international mercury reduction initiatives. Minnesota has been involved to a certain extent in all the national and regional initiatives listed in Table 14, as well as some international activities.

Mercury emissions from coal combustion in the United States are expected to decline because of technological innovation under the MACT or the Clear Skies Initiative, and that cost-effective mercury control will be adopted world wide following its development in the United States. These additional emissions reductions are expected to result in additional reductions in mercury contamination of fish after a dynamic equilibrium is reached.

Table 14 Summary of Regional, National and International Mercury Reduction Initiatives

Program	Description	Comments
Regional Initiatives		
Lake Superior Programs	Binational Program, LAMP	Virtual elimination by 2020
Great Lakes	Binational Toxics Strategy	50% reduction by 2006
St. Louis River (northern Minnesota)	Area of Concern Remedial Action Plan	
National Initiatives		
EPA's Proposed Rules for Coal-fired power plants - 2004	CAAA Section 112 Maximum Achievable Control Technology standard or Section 111 standards	68% reduction in current emissions by 2018 under Section 111 standards (15 ton emissions cap)
Mercury Legislative Proposals — Coal-fired Utility	Various proposals currently before Congress including the President's "Clear Skies Initiative"	Proposals vary; 47-90% reduction, 2008-2018 implementation.
National Voluntary Sector Initiatives	Medical, electrical, automotive, chlor-alkali	75% reduction by chlor-alkali sector, others pledged.
State Efforts	ECOS, individual states, northeastern states	Reduction goals, programs and initiatives
Product Bans (Use, sales, ingredients, etc.)	Latex paint, seed coatings, batteries, fungicides, misc.	Product bans account for substantial emissions reductions to date.
International Initiatives		
International Agreements	Aarhus (long-range transport), Basel (haz. waste), Stockholm (POPs)	All address mercury
United Nations Environment Programme	Established "Capacity Building" Mercury Program for developing countries	Concluded in 2003 that mercury is a pollutant of concern.
Commission for Environmental Cooperation	U.S., Mexico and Canada	Reduce emissions by 50% by 2006
European Union	European Community Legislation on Mercury	All member countries of the EU must implement a variety of legislated requirements.
Individual Countries	Sweden, others	Individual countries, most notably Sweden have implemented proactive mercury initiatives.

13 Public Participation

The goals of public participation include building trust, developing partnerships, encouraging creativity, facilitating learning, increasing levels of commitment, maximizing participation, and reaching consensus (Smolko *et al.*, 2002). The Agency is committed to a robust effort that meets with a wide range of stakeholders, listens to new ideas and approaches, and remains flexible.

The Agency met with key stakeholders throughout the development of the regional mercury TMDLs, including USEPA Region 5 and headquarters. The draft TMDL was available through the Agency TMDL web site, a series of public meetings were held across the state, and a 90-day formal public comment period ended on October 18, 2005. All comments received during that formal comment period, plus Agency responses, will be included in the draft TMDL package that is delivered to Region 5 EPA for their review and approval.

The Agency received more than 975 comments. The summary of the comments, and the Agency responses, can be found at http://www.pca.state.mn.us/water/tmdl/tmdl-mercuryplan.html#comments .

14 References

Atkeson, T., D. Axelrad, C. Pollman, and G. Keeler. 2002. *Integrated Summary Integrating Atmospheric Mercury Deposition and Aquatic Cycling in the Florida Everglades: An approach for conducting a Total Maximum Daily Load analysis for an atmospherically derived pollutant*. Prepared for: United States Environmental Protection Agency, October, 2002. ftp://ftp.dep.state.fl.us/pub/labs/assessment/mercury/tmdlreport.pdf

Atkinson, D.R. 2003. Air Deposition Modeling and the TMDL Program. Mercury Loadings to States and Regions. http://www.northstar.sierraclub.org/campaigns/air/mercury/epaMercuryDepositionReport2003.pdf Accessed July 29, 2004

Balogh SJ, Meyer ML, Johnson DK. 1997. Mercury and suspended sediment loadings in the lower Minnesota River. *Environmental Science and Technology* 31: 198-202.

Balogh SJ, Meyer ML, Johnson DK. 1998. Transport of mercury in three contrasting river basins. *Environmental Science and Technology* 32: 456-462.

Balogh SJ, Huang Y, Offerman HJ, Meyer ML, Johnson DK. 2002. Episodes of elevated methylmercury concentrations in prairie streams. *Environmental Science and Technology* 36: 1665-1670.

Balogh SJ, Huang Y, Offerman HJ, Meyer ML, Johnson DK. 2003. Methylmercury in rivers draining cultivated wetlands. *Science of the Total Environment.* 304: 305-313.

Barr Engineering. 2001. *Substance Flow Analysis of Mercury in Products.* Prepared for Minnesota Pollution Control Agency. August, 2001. http://www.pca.state.mn.us/air/mercury-mn.html#publications.

Bennett, J. P,. and C. M. Wetmore. 2000. 16-Year trends in elements of lichens at Theodore Roosevelt National Park, North Dakota. *Science of the Total Environment* 263:231-241.

Branfireun, B. A., Roulet, N. T., Kelly, C. A., Rudd, W. M. 1999. *In situ* sulphate stimulation of mercury methylation in a boreal peatland: toward a link between acid rain and methylmercury contamination in remote environments. *Global Biogeochemical Cycles* 13, 743-750.

Chen, C. Y. and C.L. Folt. 2005. High Plankton Densities Reduce Mercury Biomagnification. *Environmental Science and Technology* 39: 115-121.

D'Itri, F.M., C.S. Annett and A.W. Fast. 1971. Comparison of mercury levels in an oligotrophic and eutrophic Lake. *J. Mar. Technol. Soc*. 5(6): 10-14.

Dilks, D.W. and P.L. Freedman. 2004. Improved Consideration of the Margin of Safety in Total Maximum Daily Load Development. *Journal of Environmental Engineering.* 130: 690-694.

Driscoll, C.T., Blette, V., Yan, C., Schofield, C.L., Munson, R., and J. Holsapple. 1995. The role of dissolved organic carbon in the chemistry and bioavailability of mercury in remote Adriondack lakes, *Water Air and Soil Pollution* 80: 499-508.

EERC. 2001. *Fish consumpt:ion survey: Minnesota and North Dakota. Energy and Environmental Research Center*. University of North Dakota, Grand Forks.

Engstrom, D. R., and E. B. Swain. 1997. Recent declines in atmospheric mercury deposition in the Upper Midwest. *Environmental Science and Technology* 31:960-967.

Engstrom, D. R., K. Thommes, S. J. Balogh, E. B. Swain, and H. A. Post. 1999. *Trends in Atmospheric Mercury Deposition across Minnesota: Evidence from Dated Sediment Cores from 50 Minnesota Lakes. Final Report to the Legislative Commission on Minnesota Resources.*

Expert Panel on Mercury Atmospheric Processes. 1994. *Mercury Atmospheric Processes: A Synthesis Report*. Convened March 16-18, 1994 Tampa, Florida, U.S.A. EPRI/TR-104214.

Fink, L.E. 2001. Appendix 7-11: *The Effect of Surface and Pore Water Quality on Mercury Bioaccumulation*. 2001 Everglades Consolidated Report.

FTN Associates, Ltd. 2002. *TMDLs for Segments Listed for Mercury in Fish Tissue for the Ouachita River Basin, and Bayou Bartholomew, Arkansas and Louisiana to Columbia.* Prepared for USEPA Region VI, Dallas, TX. Approved December 18, 2002.

FTN Associates, Ltd. 2003. *TMDLs for Lakes Listed for Mercury in Fish Tissue for the Ouachita River Basin, Arkansas*. Prepared for USEPA Region VI, Dallas, TX. DRAFT November 20, 2003.

Gilmour, C. C., and E. A. Henry. 1991. Mercury methylation in aquatic systems affected by acid deposition. *Environmental Pollution* 71:131-169.

Gilmour, C. C., E. A. Henry, and R. Mitchell, 1992. Sulfate stimulation of mercury methylation in freshwater sediments. *Environmental Science and Technology* 26:2281-2287.

Glass, G. E., and J. A. Sorensen. 1999. Six-year trend (1990-1995) of wet mercury deposition in the Upper Midwest, U.S.A. *Environmental Science and Technology* 33:3303-3312.

Greenfield, B.K., Hrabik, T.R., Harvey, C.J., and S.R. Carpenter. 2001. Predicting mercury levels in yellow perch: use of water chemistry, trophic ecology, and spatial results. *Canadian Journal of Fisheries & Aquatic Sciences* 58: 1419-1429.

Hagen, D.A., J.W. Vincent, and P.G. Welle. 1999. Economic benefits of reducing mercury deposition in Minnesota. Conducted on behalf of the Minnesota Pollution Control Agency and the Legislative Commission on Minnesota Resources. http://www.pca.state.mn.us/publications/reports/mercury-economicbenefits.pdf

Hakanson, L. 1980. The quantitative impact of pH, bioproduction and Hg-contamination on the Hg-content of fish (pike). Environ. Pollut. (Series B) 1: 285-304.

Heiskary, S.A., C.B. Wilson, and D. P. Larsen. 1987. Analysis of regional patterns in lake water quality: using ecoregions for lake management in Minnesota. *Lake and Reservoir Management,* 3: 337-344. www.pca.state.mn.us/data/eda/wqguide.html

Heiskary, S.A., and W.W. Walker, Jr. 1988. Developing Phosphorus Criteria for Minnesota Lakes. *Lake and Reservoir Management* 4(1): 1-9.

Heiskary, S.A., and C.B. Wilson. 1989. The Regional Nature of Lake Water Quality Across Minnesota: An Analysis for Improving Resource Management. *Journal Minnesota Academy Science* 55(1): 71-77.

McCollor S, and Heiskary S. 1993. Selected water quality characteristics of minimally impacted streams from Minnesota's seven ecoregions. Minnesota Pollution Control Agency, St. Paul, MN.

Hintelmann H., R. Harris, A. Heyes, J.P. Hurley, C.A. Kelly, D.P. Krabbenhoft, S. Lindberg, J.M. Rudd, K.J. Scott, and V.L. St. Louis. 2002. Reactivity and Mobility of New and Old Mercury Deposition in a Boreal Forest Ecosystem during the First Year of the METAALICUS Study. *Environmental Science and, Technology.* 36: 5034-5040.

Hrabik, T. R., and C. J. Watras. 2002. Recent declines in mercury concentration in a freshwater fishery: isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. *The Science of the Total Environment* 297:229-237.

Hylander , L.D. and M. Meili. 2003. 500 years of mercury production: global annual inventory by region until 2000 and associated emissions. *The Science of the Total Environment* 304: 13-27.

Jackson, A. M., E. B. Swain, C. A. Andrews, and D. Rae. 2000. Minnesota's Mercury Contamination Reduction Initiative. *Fuel Processing Technology* 65-66 (2000):79-99

Jeremiason, J. D., E. B. Swain, E. A. Nater, J. B. Cotner, B. M. Johnson, D. R. Engstrom, and J. E. Almendinger. 2003. Sulfate Addition Enhances Concentrations and Export of Methylmercury from a Treated Wetland. Abstract. Presented at the American Society of Limnology and Oceanography annual meeting, Salt Lake City. February 2003.

Jeremiason, J. D., D. R. Engstrom, E. B. Swain, E. A. Nater, B. M. Johnson, J. E. Almendinger, B. A. Monson, and R. K. Kolka. 2006. Sulfate addition increases methylmercury production from an experimental wetland. *Environmental Science and Technology,* IN PRESS.

Johnels, A.G., T. Westermark, W. Berg, P.I. Persson, and B. Sjöstrand. 1967. Pike (*Esox lucius* L.) and some other aquatic organisms in Sweden as indicators of mercury contamination in the environment. *Oikos* 18: 323-333.

Kamman, N. C., and D. R. Engstrom. 2002. Historical and present fluxes of mercury to Vermont and New Hampshire lakes inferred from ^{210}Pb dated sediment cores. *Atmospheric Environment* 36:15999-1609.

Lamborg, C.H., W.F. Fitzgerald, J. O'Donnell, and T. Torgersen. 2002. A non-steady-state compartmental model of global-scale mercury biogeochemistry with interhemispheric atmospheric gradients. *Geochimica et Cosmochimica Acta* 66(7): 1105-1118.

Mason, R.P. and G. R. Sheu. 2002. Role of the ocean in the global mercury cycle. *Global Biogeochemical Cycles* 16 (4), 1093.

Mason, R.P., M.L. Abbott, R.A. Bodaly, O. R. Bullock, Jr., C.T. Driscoll, D. Evers, S.E. Lindberg, M. Murray, and E.B. Swain. 2004. Monitoring the environmental response to changing atmospheric mercury deposition. *Environmental Science & Technology* (In Review).

Minnesota Department of Health (MDH). 2004. *Eat Fish Often? A Minnesota Guide to Eating Fish.* http://www.health.state.mn.us/divs/eh/fish/index.html

Minnesota Pollution Control Agency (MPCA). 2000. *Guidelines for the development of surface water quality standards for toxic substances for protection of aquatic life, human health, and wildlife.* Environmental Outcomes Division. August 2000.

Minnesota Pollution Control Agency (MPCA). 2002. *Mercury Reduction Program: Progress Report to Minnesota Legislature*. January 2002. http://www.pca.state.mn.us/hot/legislature/reports/2002/mercury-02.pdf

Minnesota Pollution Control Agency (MPCA). 2004a. *Guidance Manual for Assessing the Quality of Minnesota Surface Waters: for the Determination of Impairment 305(b) Report and 303(d) List*. January 2004. http://www.pca.state.mn.us/publications/manuals/tmdl-guidancemanual04.pdf

Minnesota Pollution Control Agency (MPCA). 2004b. *Estimated Mercury Emissions in Minnesota for 1990, 1995, & 2000: March 2004 Update.* http://www.pca.state.mn.us/air/mercury.html

Minnesota Pollution Control Agency (MPCA). 2005. *2005 Mercury Reduction Progress Report to Minnesota Legislature: Appendix A Mercury Emissions Inventory for Minnesota.* http://www.pca.state.mn.us/publications/reports/lrp-mercury05-appa.pdf

Mugan, T. (Tom.Mugan@dnr.state.wi.us). 10 October 2005. RE: Mercury Data Summaries. E-mail to Matthew Gluckman (Gluckman.Matthew@epamail.epa.gov).

Norstrom, R. J., A.E. McKinnon and A.S.W. DeFreitas. 1976. A bioenergetics-based model for pollutant accumulation by fish. Simulation of PCB and MeHg residue levels in Ottawa River yellow perch (*Perca flavescens*). J. Fish Res. Board Can. 33: 248-267.

Norton, S. A., G. C. Evans, and J. S. Kahl. 1997. Comparison of Hg and Pb fluxes to hummocks and hollows of ombrotrophic Big Heath Bog and to nearby Sargent Mt. Pond, Maine, USA. *Water, Air, and Soil Pollution* 100:271-286.

Omernik, J.M., and A.L. Gallant. 1988. *Ecoregions of the Upper Midwest States*. EPA/600/3-88/037. Corvallis, OR: United States Environmental Protection Agency. 56 p.

Pacyna, E.G., and J.M. Pacyna. 2002. Global emissions of mercury from anthropogenic sources in 1995. *Water, Air, and Soil Pollution* 137:149-165.

Parsons. 2003. *Mercury TMDLs for Little River and Catahoula Lake Watershed: Subsegments 081601, 081602, 081603, and 081605 TMDL Report.* Prepared for USEPA Region VI, Dallas, TX. February 2003.

Pickhardt, P. C.; Folt, C. L.; Chen, C. Y.; Klaue, B.; Blum, J. D. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. *Proc. Natl. Acad. Sci. U.S.A.* **2002**, *99*, 4419-4423.

Pirrone, N., G. J. Keeler, and J. O. Nriagu. 1996. Regional differences in world-wide emissions of mercury to the atmosphere. *Atmospheric Environment* 30:2981-2987.

Pollman, C. D., and D. B. Porcella. 2002. *Trends in mercury emissions and concentrations in biota in South Florida: A critical analysis. Final report to the Florida Electric Power Coordinating Group (FCG).* 49 pp. typescript.

Rae, D. 1997. Impact of Mercury Reductions in Minnesota. Report prepared for the Minnesota Pollution Control Agency and Hagler Bailly Services, Inc., September 30, 1997.

Schuster, P.F., D.P. Krabbenhoft, et al., 2002. Atmospheric Mercury Deposition during the Last 270 Years: A Glacial Ice Core Record of Natural and Anthropogenic Sources. *Environmental Science & Technology*. 36(11):2303-2310. http://toxics.usgs.gov/pubs/FS-051-02/pdf/fs-051-02.pdf

Seigneur, C., P. Karamchandani, K. Vijayaraghavan, K. Lohman, R.-L. Shia, and L. Levin. 2003. On the effect of spatial resolution on atmospheric mercury modeling. *The Science of the Total Environment* 304:73-81.

Seigneur, C., K. Vijayaraghavan, K. Lohman, P. Karamachandani, and C. Scott. 2004. Global source attribution for mercury deposition in the United States. *Environmental Science & Technology* 38(2): 555-569.

Slemr, F., E.-G. Brunke, R. Ebinghaus, C. Temme, J. Munthe, I. Wangberg, W. Schroeder, A. Steffen, and T. Berg. 2003. Worldwide trend of atmospheric mercury since 1977. *Geophysical Research Letters* 30(10):1516

Smolko, B.A., R.R. Huberd, and N. Tam-Davis. 2002. Creating meaningful stakeholder involvement in watershed planning in Pierce County, Washington. JAWRA 38(4): 981-994.

Sorensen J.A., G.E. Glass, K.W. Schmidt, J.K. Huber, and G.R. Rapp, Jr. 1990. Airborne Mercury Deposition and Watershed Characteristics in Relation to Mercury Concentrations in Water, Sediments, Plankton, and Fish of Eighty Northern Minnesota Lakes. *Environmental Science & Technolology* 24(11): 1716-1727.

Steinnes, E., T. Berg, and T. E. Sjobakk. 2003. Temporal and spatial trends in Hg deposition monitored by moss analysis. *The Science of the Total Environment* 304:215-219.

Sunderland, E. M., and G. L. Chmura. 2000. An inventory of historical mercury emissions in maritime Canada: implications for present and future contamination. *The Science of the Total Environment* 256:39-57.

Swain, E. B., D. R. Engstrom, M. E. Brigham, T. A. Henning, and P. L. Brezonik. 1992. Increasing rates of atmospheric mercury deposition in midcontinental North America. *Science* 257:784-787.

United States Environmental Protection Agency (USEPA). 1997. *Mercury Study Report to Congress*. EPA-452/R-97-005. http://www.epa.gov/oar/mercuy.html

United States Environmental Protection Agency (USEPA). 2001a. *Mercury TMDLs for Subsegments within Mermentau and Vermilion-Teche River Basins.* January 19, 2001.

United States Environmental Protection Agency (USEPA). 2001b. *Total Maximum Daily Load (TMDL) for Total Mercury in Fish Tissue Residue in the Middle & Lower Savannah River Watershed*. February 28, 2001.

United States Environmental Protection Agency (USEPA). 2001c. *Water Quality Criterion for the Protection of Human Health: Methylmercury.* Office of Science and Technology, Washington, D.C. EPA-823-R-01-001. http://www.epa.gov/waterscience/criteria/methylmercury/

United States Environmental Protection Agency (USEPA). 2001d. *Mercury Maps: A Quantitative Spatial Link Between Air Deposition and Fish Tissue. Peer Reviewed Final Report. U.S. Environmental Protection Agency. EPA-823-R-01-009.*

United States Environmental Protection Agency (USEPA). 2004. *Press Briefing Presentation on Fish Consumption Advisories, August 24, 2004.* http://www.epa.gov/waterscience/fish/advisories/briefing.pdf

Watras, C. J., K. A. Morrison, R. J. M. Hudson, T. M. Frost, and T. K. Kratz. 2000. Decreasing mercury in northern Wisconsin: temporal patterns in bulk precipitation and a precipitation-dominated lake. *Environmental Science and Technology* 34:4051-4057.

Western Lake Superior Sanitary District (WLSSD). 1997. *Blueprint for Mercury Elimination: Mercury Reduction Project Guidance for Wastewater Treatment Plants.*

Appendix A

Impaired Waters Covered by the Statewide Mercury TMDL

Appendix B
NPDES Permits Covered by the Statewide Mercury TMDL