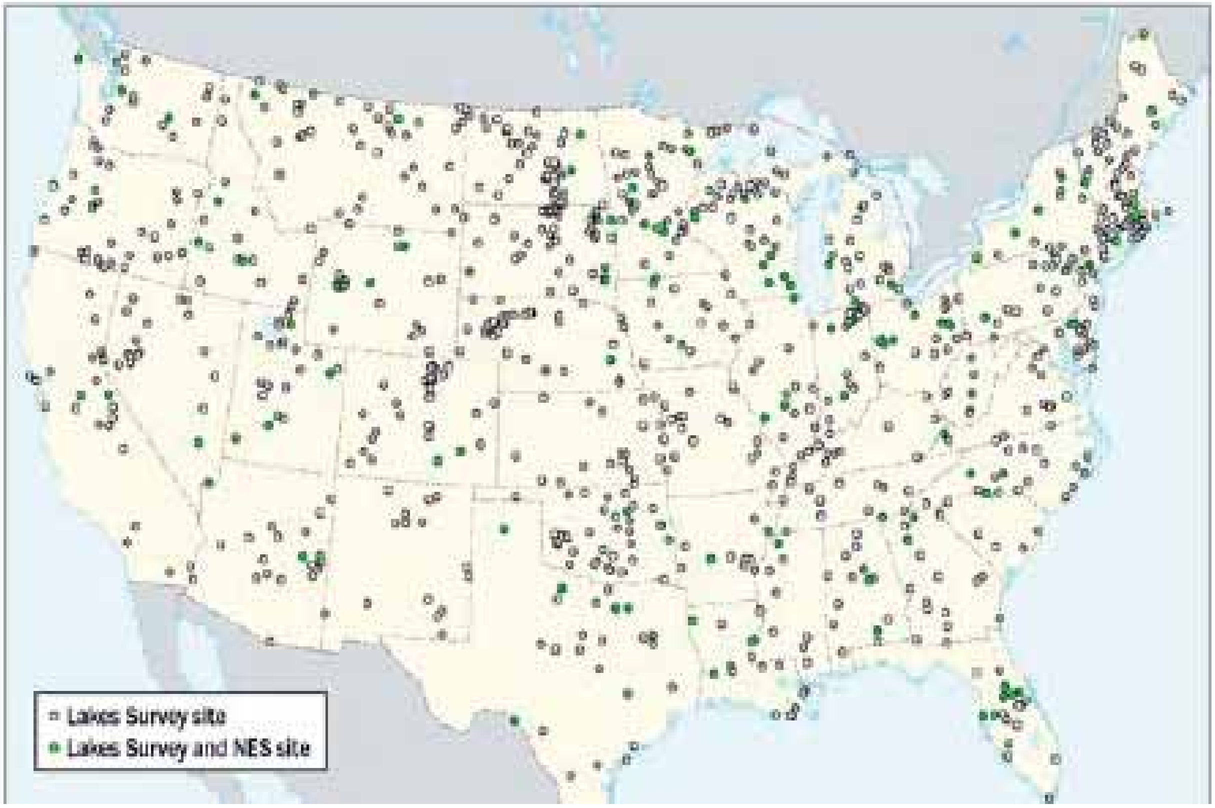

Minnesota National Lakes Assessment Project:

Water Mercury Concentrations in Minnesota Lakes

This report is part of a series based on Minnesota's participation in U.S. EPA's 2007 National Lake Assessment

October 2008

Minnesota Pollution Control Agency

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Introduction

National Lakes Assessment Project (NLAP) Overview

The U.S. Environmental Protection Agency (EPA) has a responsibility to assess the health of the nation's water resources. One of the methods for assessment is a statistically based survey. The Survey of the Nation's Lakes, conducted in 2007, is one of a series of water surveys being conducted by states, tribes, EPA, and other partners. In addition to lakes, partners will also study coastal waters, wadable streams, rivers, and wetlands in a revolving sequence. The purpose of these surveys is to generate statistically valid and environmentally relevant reports on the condition of the nation's streams, lakes, wetlands, and estuaries at nation-wide and regional scales.

The goal of the Lakes Survey is to address two key questions about the quality of the Nation's lakes, ponds, and reservoirs:

- What percent of the Nation's lakes are in good, fair, and poor condition for key indicators of trophic state, ecological health, and recreation?
- What is the relative importance of key stressors such as nutrients and pathogens?

The sampling design for this survey is a probability-based network that provides statistically valid estimates of the condition of all lakes, with a known degree of confidence. Sample sites are selected at random to represent the condition of all lakes across the nation and each region. A total of 909 lakes in the conterminous U.S. are included in the Lakes Survey. The sample set is comprised of natural and built freshwater lakes, ponds, and reservoirs greater than 10 acres and at least one meter in depth located in the conterminous United States.

The typical sampling effort at each site includes a variety of samples and measurements collected at a mid-lake index site, which is often at the deepest point in the lake. Samples include a two meter integrated sample for water chemistry, chlorophyll-a, microcystin and algal identification; oxygen and temperature profiles; zooplankton tow; and sediment core sample for diatom reconstruction of total phosphorus (based on top and bottom slices from the core) and surface sediment sample for mercury. In addition, 10 random near-shore sites are qualitatively assessed for various littoral and riparian habitat-related measures and a sample for a bacterial indicator was collected. Further details on the survey including methods, parameters measured, and statistical design may be found on the EPA NLAP web page at: http://www.epa.gov/owow/lakes/lakessurvey/.

Minnesota's NLAP Overview

Minnesota's 2007 NLAP effort was led by the Minnesota Pollution Control Agency (MPCA) and Minnesota Department of Natural Resources (MDNR). Various other collaborators were engaged in this study as well including the U.S. Forest Service (USFS), Minnesota Department of Agriculture (MDA), and U.S. Geological Survey (USGS). MPCA and MDNR cooperated on initial planning of the survey and conducted a vast majority of the sampling, which took place in July and August for most lakes. USFS staff sampled remote lakes in the Boundary Waters Canoe Area Wilderness (BWCAW).

Minnesota received 41 lakes as a part of the original draw of lakes for the national survey–the most of any of the lower 48 states. Minnesota chose to add nine lakes to the survey to yield the 50 lakes needed for statistically-based statewide estimates of condition (Figure 1). In addition to the 50 lakes, several reference lakes were later selected and sampled by USEPA as a part of the overall NLAP effort. Data from the reference lakes provide an additional basis for assessing lake condition as a part of NLAP.

In addition to adding lakes, Minnesota chose to add several value-added measurements to the survey of lakes. The additional data collection included: pesticide samples (in conjunction with the MDA); water mercury (in conjunction with USGS); sediment samples for analysis of metals, trace organics and other parameters; identification of macrophytes and maximum rooting depth of macrophytes at the random near-shore sites; and samples for microcystin at the index and a random near-shore site. Each of these add-ons and several of the standard assessments is the subject of a series of reports that draw from the NLAP work.

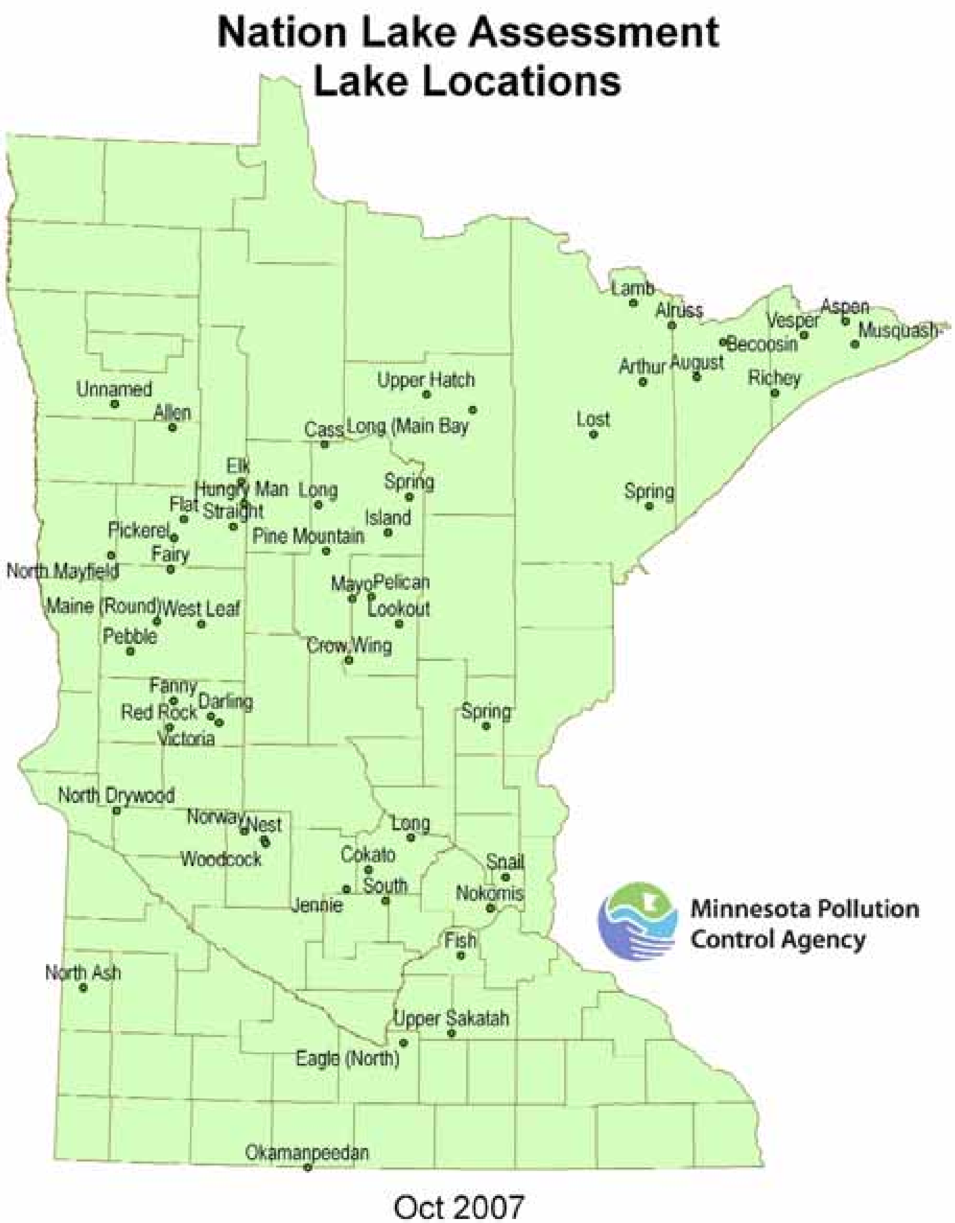

Figure 1 Location of Minnesota's NLAP lakes as surveyed in 2007

Report Focus: Water Mercury (Hg) Concentrations in Minnesota Lakes

Background on Mercury in Minnesota

Humans and wildlife are primarily exposed to mercury from the consumption of fish. Minnesota has a four-decade record of mercury in fish from lakes and rivers throughout the state and has the most extensive fish consumption advisory in the United States. Fish are excellent indicators of mercury levels in a waterway because they integrate the mercury over the life of the fish; however, mercury levels in water are important in showing natural variability and responsiveness to changes in mercury loading to watersheds. Mercury in water is considerably less well known, mostly because the concentrations are extremely low in water, requiring special sampling and analytical techniques. Mercury concentrations in fish are 1 million to 10 million times higher than in water and therefore fish analysis for mercury requires less sensitive techniques than water analysis. Despite the greater difficulty of obtaining mercury concentrations in water, it is important to gather the data because it is useful to build an understanding of how the mercury content of fish is related to the water in which the fish grow. Minnesota currently has a water quality standard for mercury in water of 6.9 ng/L (with a lower standard of 1.3 ng/L in the Lake Superior Basin, as adopted for waters within the Great Lakes basin by each bordering state). In 2008 the state promulgated a water quality standard for methylmercury in fish tissue (0.2 mg/kg). To translate the new fish tissue-based water quality standard to a water concentration, a bioaccumulation factor (BAF) would be necessary, which is the ratio of mercury concentrations in fish and water.

Mercury concentrations in lake water have been collected as part of temporary projects rather than routine monitoring. Gary Glass, USEPA, and John Sorensen, University of Minnesota – Duluth, collected mercury samples from water, zooplankton, and fish from 80 lakes in northeast Minnesota (Sorensen et al. 1990). From 1992 to 1994, the Acid Precipitation program at the MPCA seasonally monitored 12 low alkalinity lakes in northern Minnesota, which included sample collection for total and methylmercury in water and zooplankton (Monson and Brezonik 1998). An intensive five year study of water, soil, invertebrates, and fish in 17 interior lakes of Voyageurs National Park in northern Minnesota (Wiener et al. 2006). Other data have been collected as part of environmental review and as part of the routine lake monitoring program. Generally the additional data collection projects included 10-15 lakes sampled at least three times (spring, summer, and fall) per year for one to two years. None of these previous data collection efforts were probability based site selection. Nevertheless, they provide a basis for comparison with the NLAP Hg results.

The purpose of the mercury data collection and analysis was to provide unbiased estimates of water mercury concentrations (total and methyl) for Minnesota and other Midwestern states. Hypotheses being pursued based on the NLAP and related datasets are as follows:

- How does NLAP distribution of values compare to previous estimates?
- What are the spatial patterns in water mercury?
- What is the relationship among water mercury concentrations and other physicochemical factors (e.g. dissolved organic carbon)

Methods

All lakes were sampled once, except Pebble (Lake ID: 56082900) and South (Lake ID: 43001400). The second sampling from these two lakes was excluded for statistical analysis so that all lakes were represented by a single sample event collected in July or August of 2007.

The US Geological Survey Mercury Research Laboratory in Middleton, Wisconsin, provided the sample bottles and analyzed the water samples for NLAP lakes in Illinois, Minnesota, and Wisconsin. The lab also analyzed water samples for dissolved organic carbon (DOC). The laboratory is a state-of-the-art analytical facility for low level speciation of mercury, directed by Dr. D. Krabbenhoft.

Sample acquisition followed ultra-clean protocols, referred to as “Clean Hands – Dirty Hands” and is prescribed in US EPA Method 1669. Methods for sample collection, storage, and analysis of total mercury and methylmercury are described in two US Geological Survey documents (Dewild et al. 2002; Olson and Dewild 1999).

Descriptive and inferential statistics were performed with SYSTAT 12 (SYSTAT Software, Inc., 2007).

Results and Discussion

Summary Statistics

NLAP lakes have a broad distribution of surface areas and dissolved organic carbon (DOC) concentrations (Table 1). Lake areas range from 11 acres to 15,958 acres. DOC concentrations range from 4.2 ppm-C to 25.7 ppm-C. Median total mercury (THg) concentration is 0.53 ng/L, with a range of 0.16 – 5.24 ng/L. A previously sampled set of 102 Minnesota lakes, located throughout the state but nonrandomly selected, had a median THg of 1.11 ng/L and a range of 0.27-12.43 ng/L (unpublished MPCA data). Thus, the NLAP lakes as a set have lower THg than the nonrandom set of lakes; however, the comparison is problematic because the values from the nonrandom set are lakes are averages for usually multiple samples per lake (1 – 17 per lake), samples were collected before 2007, and the THg analysis was done by a different lab. Median methylmercury (MeHg) concentrations in the NLAP lakes is 0.053 ng/L, with a range of 0.006 – 0.522 ng/L. A previously sampled nonrandom set of 37 lakes had a median MeHg concentration of 0.054 ng/L, with a range of 0.013 – 0.260 ng/L. The MeHg medians are, therefore, very similar, while the NLAP lakes have a broader range of MeHg concentrations.

Table 1 Statistical summary of unfiltered total mercury (THg), unfiltered methylmercury (MeHg), methyl mercury fraction (MeHg/THg x 100), and dissolved organic carbon (DOC) for the NLAP lakes in 2007

Statistic	Lake Area (ac)	DOC (ppm C)	THg (ng/L)	MeHg (ng/L)	MeHg Fraction (%)
N of Cases	54	51	53	51	50
Arithmetic Mean	**822**	**11.8**	**0.875**	**0.093**	**11.8**
Standard Deviation	2,423	5.2	0.926	0.105	10.9
Minimum	**11**	**4.2**	**0.162**	**0.006**	**1.4**
25th percentile	87	8.1	0.313	0.022	5.1
50th percentile (median)	**174**	**10.4**	**0.526**	**0.053**	**9.2**
75th percentile	417	14.8	1.123	0.121	15.9
Maximum	**15,958**	**27.7**	**5.242**	**0.522**	**63.0**

THg and MeHg concentrations are combined to give the MeHg fraction (MeHg/THg), which is referred to as "methylation efficiency" because it indicates the efficiency of conversion of THg to MeHg in a particular system. The MeHg fraction is typically 5-10% in lakes and that agrees with the median of 9.2% in the NLAP set of lakes. The 63% MeHg fraction in Victoria Lake (Lake ID 21005400) is an outlier, despite the reasonable concentrations of THg (0.216 ng/L) and MeHg (0.136 ng/L) (see Appendix A). Unnamed Lake (Lake ID 60030700) also has a very high MeHg fraction of 41%, with THg of 0.892 ng/L and MeHg of 0.368 ng/L. There are another six lakes with MeHg fraction between 20% and 25%. These lakes need additional examination to see if the lakes or their watersheds are wetland-dominated or have physical characteristics that promote anoxic conditions necessary for methylation of mercury.

In a normal distribution, means and medians are equal. A comparison of arithmetic means and medians indicates the distributions of THg, MeHg, and lake area are skewed to the left (medians < arithmetic means). As density plots demonstrate (Figure 2), the distribution of DOC concentrations is nearly symmetrical, whereas THg, MeHg, and lake area are strongly skewed to the left. Logarithmic transformation of THg, MeHg, and lake area improved the symmetry of the distributions (Figure 3); therefore, the log-transformed variables were used for inferential statistical analysis (e.g., bivariate plots and regression).

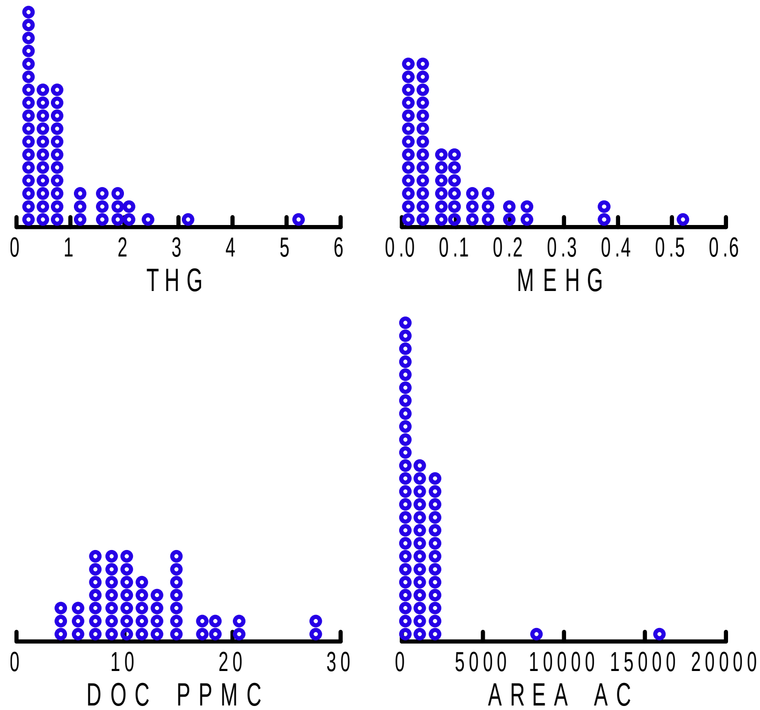

Figure 2 Density plots of THg, MeHg, DOC, and Lake Area

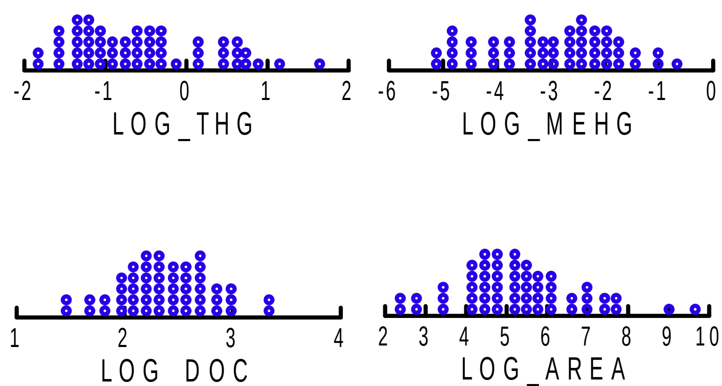

Figure 3 Density plots of Log-transformed THg, MeHg, DOC, and Area

Relationships Among Variables

The four key variables are compared in a scatterplot matrix (Figure 4), which compares all variables in bivariate plots, along with a LOWESS (locally weight least squares) smoother through the data and a histogram of the data distributions. Positive relationships are evident for THg–MeHg, THg–DOC, and MeHg–DOC. There appears to be a negative relationship between lake area and DOC, implying that the water of smaller lakes is more affected by sources of DOC such as wetlands. Pearson correlation coefficients support these graphical observations (Table 2). An ordinary least squares regression of log-transformed) THg and DOC has an $r^2 = 0.42$ and is highly significant ($p < 0.001$). The regression for MeHg and DOC has an $r^2 = 0.33$ and is highly significant ($p < 0.001$), too.

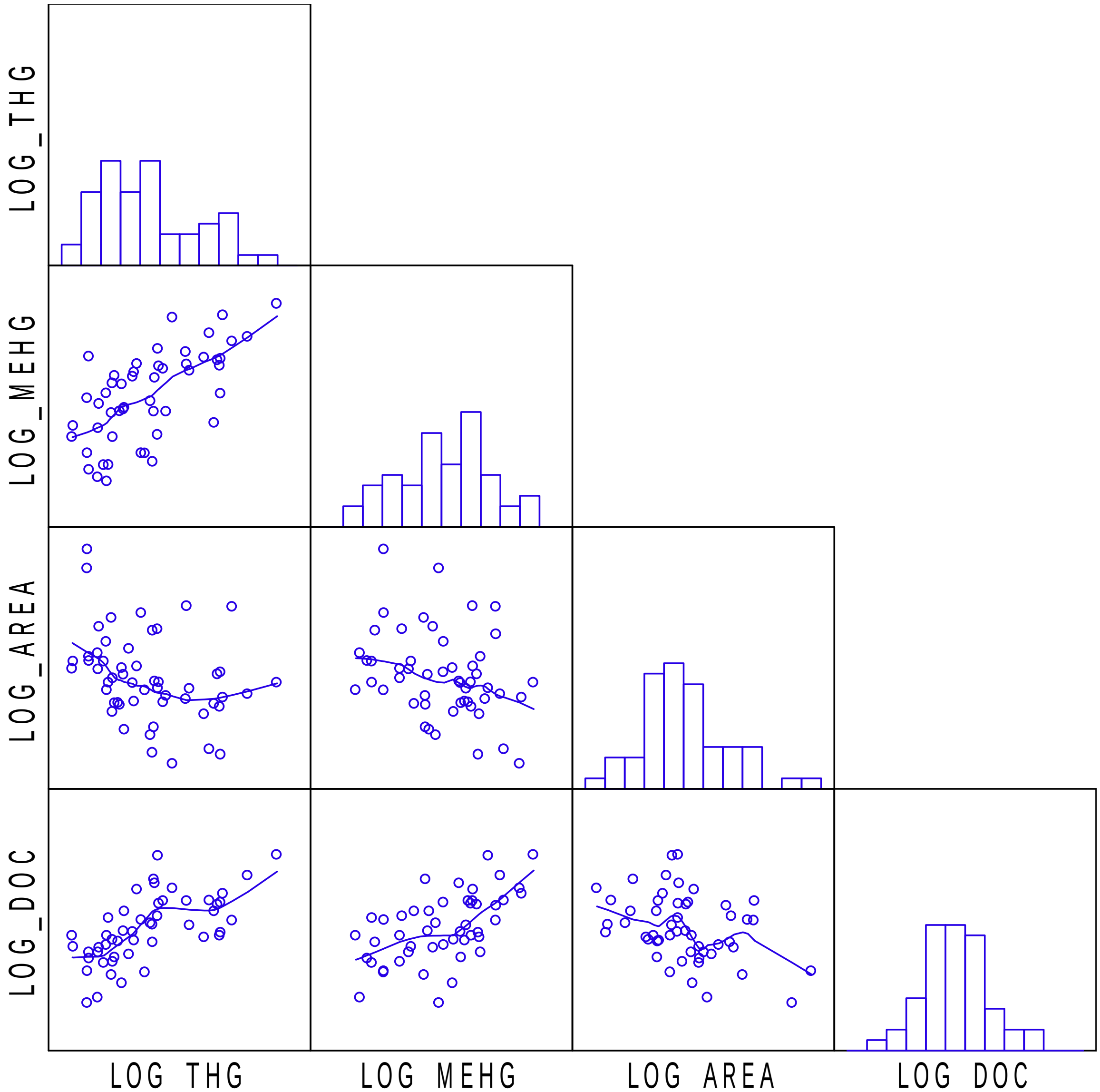

Figure 4 Scatterplot matrix of log-transformed THg, MeHg, DOC, and Area, with LOWESS smoother and histogram of data distributions

Table 2 Pearson correlation matrix of log-transformed THg, MeHg, DOC, and Area

	LOG_THG	LOG_MEHG	LOG_AREA	LOG_DOC
LOG_THG	1.000			
LOG_MEHG	0.657	1.000		
LOG_AREA	-0.330	-0.320	1.000	
LOG_DOC	0.649	0.573	-0.387	1.000

The positive relationship between MeHg and THg is somewhat surprising because they are often not correlated in lakes, rivers, and wetlands. The strong positive relationship between THg and MeHg with DOC has been observed in other lakes (Driscoll et al. 1995; Watras et al. 1995b). DOC is associated with wetland drainage (Gergel et al. 1999) and much of the Hg is methylated in wetlands by sulfate reducing bacteria (Gilmour and Henry 1992); MeHg and Hg are transported to lakes sorbed to DOC (Ravichandran 2004). In addition to the MeHg from wetlands, MeHg can also be formed within the anaerobic hypolimnion of lakes and lake sediments (Watras et al. 1995a; Watras et al. 2005). Victoria Lake, on the eastern edge of Alexandria, Minnesota, has steep shoreline slopes and a maximum depth of 60 feet, suggesting that the high MeHg fraction could be caused by internally produced MeHg rather than wetland contributions from the watershed.

Spatial Distributions

Spatial distributions of THg, MeHg, and MeHg fraction (MeHg/THg) categorized into three concentration levels—low, medium, and high—indicate that high concentrations of THg and MeHg are distributed throughout the state (Figure 5). Northeastern region of the NLF ecoregion has higher THg and MeHg concentrations compared to the southwestern region of the NLF; although the MeHg fraction may actually be somewhat higher in the southwestern NLF. Otherwise, high THg and MeHg concentrations are distributed throughout the range of NLAP lakes. THg and MeHg concentrations can vary by an order of magnitude among adjacent lake watersheds, as shown in the Voyageurs National Park (Wiener et al. 2006). As already indicated, the dominance of wetlands in a watershed hydrologically-connected to a lake has a substantial influence on the THg and MeHg concentrations.

Summary and Conclusions

THg and MeHg concentrations in the NLAP lakes are positively correlated to DOC and negatively correlated to lake surface area. The lakes in the northeastern corner of the state have consistently high THg concentrations and many have high MeHg concentrations. Elsewhere in the state, the spatial distribution does not show apparent patterns. Follow-up data analysis should compare the land cover characteristics, especially connected wetlands, within in each lake watershed to the THg and MeHg concentrations. An acceptable comparison of mercury concentrations in water and fish is not possible, because only 16 of the lakes currently have mercury concentrations in standard length northern pike or walleye (the indicator species for mercury trends in Minnesota), and only 7 of the lakes have fish-Hg data collected after 1995. Therefore, additional fish collection and tissue analysis in the NLAP lakes would be beneficial for comparison to THg and MeHg concentrations.

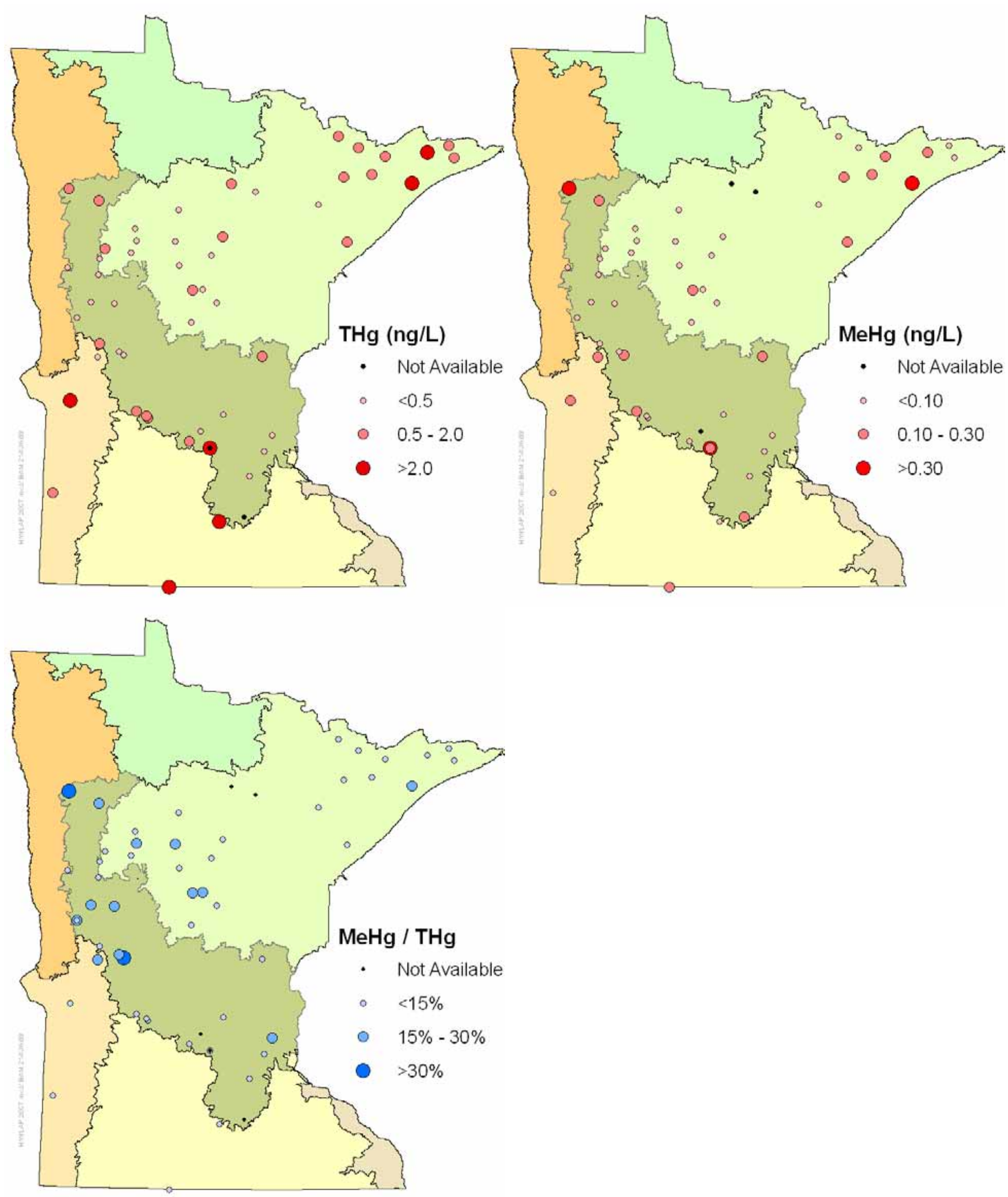

Figure 5 Spatial distributions of THg and MeHg concentrations, and MeHg fraction, overlying ecoregions

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Appendix A THg, MeHg, DOC, UV Absorbance at 254 nm, and SUVA for All Sites

Lake Name	Lake ID	EPA_SITEID	Eco-region	County	Area (ac)	Sample Date	THg (ng/L)	MeHg (ng/L)	% MeHg/ THg	DOC (mg/L)	UV-Abs (254nm)	SUVA
Allen	44015700	NLA06608-0211	NCHF	Mahnomen	143	8/7/07	0.696	0.166	24%	27.40	0.405	1.1
Alruss	69000500	NLA06608-1102	NLF	St. Louis	29	7/16/07	0.615	0.044	7%	11.60	0.076	0.7
Arthur	69015400	NLA06608-1342	NLF	St. Louis	76	7/17/07	1.989	0.108	5%	9.90	0.190	1.9
Aspen	16020400	NLA06608-0890	NLF	Cook	141	7/30/07	1.193	0.095	8%	11.30	0.311	2.8
August	38069100	NLA06608-1038	NLF	Lake	229	8/3/07	1.916	0.124	6%	14.70	0.456	3.1
Becoosin	38047200	NLA06608-0526	NLF	Lake	59	7/18/07	1.528	0.133	9%	9.70	0.172	1.8
Cass	4003000	NLA06608-0403	NLF	Beltrami	15958	8/6/07	0.211	0.012	6%	6.30	0.096	1.5
Cokato	86026300	NLA06608-0551	NCHF	Wright	545	8/20/07	0.427			7.80	0.160	2.1
Crow Wing	18015500	NLA06608-0990	NLF	Crow Wing	355	7/30/07	0.166	0.023	14%	8.60	0.139	1.6
Darling	21008000	NLA06608-1390	NCHF	Douglas	1156	7/19/07	0.257	0.041	16%	8.50	0.111	1.3
Eagle (North)	7006001	NLA06608-0279	WCBP	Blue Earth	246	8/8/07	2.019	0.053	3%	15.10		
Elk (Geraldine)	15001000	MN:15-0010	NLF	Clearwater	271	8/9/07	0.253	0.022	9%	8.00	0.126	1.6
Fairy	56035600	NLA06608-1326	NCHF	Otter Tail	134	8/6/07	0.293	0.006	2%	9.90	0.126	1.3
Fanny	21033600	NLA06608-0686	NCHF	Douglas	38	7/11/07	0.651	0.034	5%	20.30	0.540	2.7
Fish	70006900	NLA06608-0935	NCHF	Scott	173	8/6/07	0.302	0.009	3%	12.40	0.146	1.2
Flat	3024200	NLA06608-1134	NLF	Becker	1838	8/7/07	0.526	0.012	2%	12.10	0.172	1.4
Hungry Man	3002900	MN:03-0029	NLF	Becker	91	8/8/07	0.466	0.091	20%	9.30	0.133	1.4
Island	11010200	MN:11-0102	NLF	Cass	277	8/11/07	0.162	0.018	11%	9.90	0.188	1.9
Jennie	47001500	NLA06608-1175	NCHF	Meeker	1064	8/21/07	0.693	0.019	3%	12.70	0.196	1.5
Lamb	69034100	NLA06608-1150	NLF	St. Louis	110	7/17/07	0.801	0.033	4%			
Long	86006900	NLA06608-0743	NCHF	Wright	361	6/26/07	0.217	0.008	4%	7.40	0.139	1.9
Long	11048000	NLA06608-0942	NLF	Cass	284	7/10/07	0.378	0.067	18%	5.40	0.061	1.1
Long (Main Bay)	31026601	NLA06608-0958	NLF	Itasca	87	8/15/07	0.355			9.20	0.201	2.2
Lookout	18012300	NLA06608-0771	NLF	Crow Wing	226	8/13/07	0.388	0.035	9%	10.50	0.181	1.7
Lost (Horseshoe)	69061100	NLA06608-0318	NLF	St. Louis	81	7/20/07	0.366	0.034	9%			
Maine (Round)	56047600	NLA06608-1198	NCHF	Otter Tail	86	7/25/07	0.335	0.083	25%	7.50	0.094	1.3
Mayo	18040800	NLA06608-1262	NLF	Crow Wing	175	7/9/07	0.709	0.106	15%	14.90	0.415	2.8
Musquash	16010400	NLA06608-1274	NLF	Cook	133	8/1/07	0.560	0.012	2%	6.20	0.098	1.6
Nest	34015400	NLA06608-0215	NCHF	Kandiyohi	1008	8/22/07	0.638	0.009	1%	9.10	0.162	1.8
Nokomis	27001900	NLA06608-0679	NCHF	Hennepin	201	6/27/07	0.324	0.017	5%	7.10	0.099	1.4
North Ash	41005500	NLA06608-1111	NGP	Lincoln	84	7/10/07	1.812	0.025	1%	13.50	0.255	1.9
North Drywood	76016900	NLA06608-0871	NGP	Swift	117	7/12/07	3.197	0.225	7%	21.30	0.402	1.9
North Mayfield	14002900	NLA06608-0558	NCHF	Clay	35	8/9/07	0.395	0.037	9%	13.50	0.211	1.6
Norway	34025100	NLA06608-1383	NCHF	Kandiyohi	2319	8/22/07	1.138	0.112	10%	15.40	0.215	1.4
Okamanpeedan	46005100	NLA06608-0759	WCBP	Martin	2268	8/7/07	2.454	0.201	8%	12.00	0.220	1.8
Pebble	56082900	NLA06608-0174	NCHF	Otter Tail	170	7/25/07	0.455	0.082	18%	10.40	0.116	1.1
Pebble	56082900	NLA06608-0174	NCHF	Otter Tail	170	8/28/07	0.465	0.015	3%	10.10	0.114	1.1

Lake Name	Lake ID	EPA_SITEID	Eco-region	County	Area (ac)	Sample Date	THg (ng/L)	MeHg (ng/L)	% MeHg/ THg	DOC (mg/L)	UV-Abs (254nm)	SUVA
Pelican	18030800	NLA06608-0238	NLF	Crow Wing	8367	7/11/07	0.210	0.047	22%	4.20	0.035	0.9
Pickerel	3028700	NLA06608-0110	NLF	Becker	353	7/31/07	0.278	0.009	3%	7.00	0.082	1.2
Pine Mountain	11041100	NLA06608-0494	NLF	Cass	1558	7/11/07	0.317	0.032	10%	6.00	0.106	1.8
Red Rock	21029100	NLA06608-1454	NCHF	Douglas	300	7/25/07	0.489	0.113	23%	17.80	0.274	1.5
Richey	16064300	NLA06608-1018	NLF	Cook	104	8/2/07	2.103	0.389	19%	16.90	0.263	1.6
Snail	62007300	NLA06608-1447	NCHF	Ramsey	64	7/18/07	0.322	0.069	21%	9.40	0.156	1.7
South	43001400	NLA06608-0167	NCHF	McLeod	173	7/23/07	5.242	0.522	10%	27.70	0.510	1.8
South	43001400	NLA06608-0167	NCHF	McLeod	173	8/29/07		0.183		25.30	0.496	2.0
Spring	33002700	NLA06608-1283	NCHF	Kanabec	18	6/27/07	1.672	0.247	15%	15.50	0.597	3.9
Spring	69012900	NLA06608-1347	NLF	St. Louis	99	6/28/07	1.119	0.153	14%			
Spring	11002200	ELS:2D3-008	NLF	Cass	89	8/10/07	0.762	0.099	13%	15.40	0.418	2.7
Straight	3001000	NLA06608-0366	NLF	Becker	470	7/31/07	0.251	0.006	3%	4.50	0.063	1.4
Unnamed	60030700	NLA06608-0915	NCHF	Polk	11	8/8/07	0.892	0.368	41%	18.10	0.444	2.5
Upper Hatch	31077000	NLA06608-0190	NLF	Itasca	16	8/14/07	0.635			11.40	0.233	2.0
Upper Sakatah	40000200	NLA06608-1303	NCHF	Le Sueur	892	8/8/07		0.202		14.50	0.311	2.2
Vesper	16041400	NLA06608-0782	NLF	Cook	15	7/31/07	2.020	0.129	6%	10.30	0.274	2.7
Victoria	21005400	NLA06608-0622	NCHF	Douglas	417	7/24/07	0.216	0.136	63%	8.00	0.136	1.7
West Leaf	56011400	NLA06608-0878	NCHF	Otter Tail	693	8/1/07	0.290	0.053	18%	8.80	0.131	1.5
Woodcock	34014100	NLA06608-1239	NCHF	Kandiyohi	180	7/24/07	0.662	0.079	12%	19.30	0.238	1.2

Ecoregions: North Central Hardwood Forest (NCHF); Northern Glaciated Plains (NGP); Northern Lakes and Forests (NLF); Western Corn Belt Plain (WCBP)

SUVA = UV254nm x 100 / DOC

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