



Distribution Patterns of Mercury in Lakes and Rivers of Northeastern North America

IAN F. DENNIS,¹ THOMAS A. CLAIR,^{1,*} CHARLES T. DRISCOLL,² NEIL KAMMAN,³ ANN CHALMERS,⁴
JAMIE SHANLEY,⁴ STEPHEN A. NORTON⁵ AND STEVE KAHL^{6,7}

¹*Environment Canada, Atlantic Region, PO Box 6227, Sackville, NB E4L 1G6, Canada*

²*Department of Chemistry, Syracuse University, Center for Science and Technology Syracuse, New York 13244-4100, USA*

³*VT Department of Environmental Conservation, 103 S Main 10N, Waterbury, VT 05671, USA*

⁴*U.S. Geological Survey, PO Box 628, Montpelier, Vermont 05601, USA*

⁵*Department of Earth Sciences, University of Maine, Orono, Maine 04469-5790, USA*

⁶*George Mitchell Center, University of Maine, Orono, Maine 04469, USA*

⁷*Center for the Environment, Plymouth State University, Plymouth, NH USA*

Accepted 4 December 2004

Abstract. We assembled 831 data points for total mercury (Hg_t) and 277 overlapping points for methyl mercury (CH_3Hg^+) in surface waters from Massachusetts, USA to the Island of Newfoundland, Canada from State, Provincial, and Federal government databases. These geographically indexed values were used to determine: (a) if large-scale spatial distribution patterns existed and (b) whether there were significant relationships between the two main forms of aquatic Hg as well as with total organic carbon (TOC), a well know complexer of metals. We analyzed the catchments where samples were collected using a Geographical Information System (GIS) approach, calculating catchment sizes, mean slope, and mean wetness index. Our results show two main spatial distribution patterns. We detected loci of high Hg_t values near urbanized regions of Boston MA and Portland ME. However, except for one unexplained exception, the highest Hg_t and CH_3Hg^+ concentrations were located in regions far from obvious point sources. These correlated to topographically flat (and thus wet) areas that we relate to wetland abundances. We show that aquatic Hg_t and CH_3Hg^+ concentrations are generally well correlated with TOC and with each other. Over the region, CH_3Hg^+ concentrations are typically approximately 15% of Hg_t . There is an exception in the Boston region where CH_3Hg^+ is low compared to the high Hg_t values. This is probably due to the proximity of point sources of inorganic Hg and a lack of wetlands. We also attempted to predict Hg concentrations in water with statistical models using catchment features as variables. We were only able to produce statistically significant predictive models in some parts of regions due to the lack of suitable digital information, and because data ranges in some regions were too narrow for meaningful regression analyses.

Keywords: total mercury; methyl mercury; freshwater; northeastern North America; GIS/landscape analysis

*To whom correspondence should be addressed:
Tel.: + 506-364-5070; Fax: + 506-364-5062;
E-mail: tom.clair@ec.gc.ca

Introduction

Miller et al. (this issue) and Van Arsdale (this issue), demonstrate that mercury (Hg) is ubiquitous in the atmosphere of northeastern North America. The deposition pattern in the region is determined by large-scale factors such as orographic and vegetational characteristics of specific areas, as well as by local point sources in more developed areas (Miller et al., 2005). After Hg deposition in either wet or dry form, a number of factors influence Hg concentrations in surface waters. In this work, we study patterns of surface water Hg to see if they parallel the same large-scale patterns as shown by deposition.

Although there are large numbers of lake, catchment, or locally-specific Hg studies, there are few that evaluate freshwater Hg distribution on a large scale. Krabbenhoft et al. (2002) sampled 90 high altitude lakes in the western US. They noted a weak, but significant correlation between total Hg and DOC. They also showed a stronger correlation between total Hg (Hg_t) and methyl Hg (CH_3Hg^+), suggesting that Hg loading was the main determinant of CH_3Hg^+ concentrations. Rice (1999) found higher Hg levels in stream sediments in and near urban areas across the US. Landis et al. (2002) showed that local Hg emissions significantly influence surface water concentrations in the Chicago, IL region.

Factors other than atmospheric loading have also been shown to control surface water Hg concentrations. Driscoll et al. (1995), Clair and Ehrman (1998), and Lu and Jaffe (2001) showed that DOC, which is mostly controlled by the presence of wetlands in catchments, was correlated positively with Hg_t and CH_3Hg^+ in water.

Clearly, several factors are important in influencing Hg concentrations in the surface waters of the Northeast. As part of the Northeastern Ecosystem Research Cooperative (NERC) Hg study, we assembled surface water data from 831 lakes from across northeastern North America to assess patterns in surface water Hg concentrations across the region. The main objective was to assess the relationships between Hg concentrations in water and terrain parameters especially percentage of wetlands, or determine whether other factors such as urbanization were as important.

Our data also allowed us to see if the statistical relationships between DOC and CH_3Hg^+ and Hg_t (Driscoll et al., 1995, Lu and Jaffe, 2001) applied over the whole region, or whether other relationships existed. Finally, we produced maps showing large-scale patterns of surface water Hg_t and CH_3Hg^+ for a portion of the North American continent that included both heavily populated regions, such as the US northeast, and the very lightly populated Island of Newfoundland.

We identified two possible factors controlling Hg distribution in freshwaters of the region. The first was that distance from the heavily populated New York–Boston–Portland, USA corridor would be an important determinant of Hg_t and CH_3Hg^+ concentrations in surface waters. The second factor was that geographical features of the landscape, especially wetlands could instead be used to predict dissolved Hg concentrations in regions away from the heavily populated ones.

Data and methods

Numbers of samples and the jurisdictions where they were collected are identified in Table 1. Surface water sites where Hg_t and CH_3Hg^+ samples were collected are shown in Fig. 1. Specific descriptions, locations, water body type and sizes for the 831 sites used in the analysis are available at the project web site (Briloon, 2004). More than 95% of the sites were lakes with measurable catchments, although a few stream sites were included where no lake data were available.

All Hg_t samples used were unfiltered and thus provide an estimate of particulate plus dissolved Hg_t and CH_3Hg^+ . This approach was taken as all contributors had this type of data available, while few had filtered samples. All samples were collected using ultraclean methods. Specific procedures followed by the individual contributors can be acquired from the project website (Briloon, 2004). Contributors insured that their data were properly sampled, analyzed and data were quality controlled. All data were submitted in a common format for statistical analysis.

In this work, we decided to only use data from spatial surveys collected during base flow periods to provide a consistent “snapshot” of conditions

Table 1. State/Provincial statistics from the database used

Region	Hg _t					n	CH ₃ Hg ⁺					n
	Max	Min	Mean (ng l ⁻¹)	Stdev.	Median		Max	Min	Mean (ng l ⁻¹)	Stdev.	Median	
All	19.50	ND	2.72	2.25	2.09	831	3.12	0.01	0.30	0.36	0.22	277
MA	13.68	0.53	3.54	2.96	2.61	29	1.82	0.06	0.32	0.37	0.25	29
ME	7.01	0.09	1.97	1.34	1.56	120	1.04	0.05	0.31	0.27	0.19	18
NB	9.26	ND	2.90	2.02	2.37	40	ND	ND	ND	ND	ND	ND
NF	7.86	0.11	2.95	1.43	2.82	43	ND	ND	ND	ND	ND	ND
NH	9.66	0.40	1.89	1.53	1.46	85	3.12	0.04	0.33	0.36	0.28	85
NS	19.50	0.05	3.54	2.60	2.99	365	3.02	0.13	0.59	0.69	0.31	24
NY	7.01	1.36	3.24	0.26	3.00	32	0.96	0.03	0.33	0.26	0.24	32
VT	7.57	0.09	1.45	1.13	1.18	117	0.70	0.01	0.20	0.14	0.18	89

across the whole region. Hg seasonal changes have been discussed by Shanley et al. (2002, this issue) and often show that extreme values commonly occur during winter and early spring during runoff. Hydrological extreme events obscured landscape patterns of Hg concentrations. Consequently, we removed them from our analysis.

Hg_t data were collected from much of northeastern North America (Fig. 1, top). Sample coverage of Vermont (VT), New Hampshire (NH) and southern Maine (ME) was very good. Nova Scotia (NS) and Newfoundland (NF) sites are relatively well represented. Eastern Massachusetts (MA), as well as the Adirondack Mountain region of northern New York (NY) and southern New Brunswick (NB) are also well covered. A few sites were sampled on the NY side of Lake Champlain at the NY-VT border and were assigned to the VT group. There were fewer CH₃Hg⁺ data available for our analysis (Fig. 1b). Very good coverage of CH₃Hg⁺ data was available for southern New England and the Adirondack Mountains. We had few sites in ME and NS and no coverage for NB and NF.

Data analysis was done following political boundaries because these also tended to represent different physiographic regions, as well as differing sampling programs. VT, NH, and NY sites were generally located in hilly regions, MA in flatter terrain, whereas ME, NB, NS, and NF sites were in more mixed topographies. As mapping information was also more readily available by political boundaries, our interpretation of Hg data was made along these units. We related our study region to the Washington, DC-Boston MA-Port-

land ME urban corridor, which we call the High Population Corridor (HPC) (Fig. 1, top).

We developed isopleth maps from the Hg_t and CH₃Hg⁺ data to provide a spatial view of their distribution. We mapped using ESRI ArcView GIS 3.2 software. Isopleth calculations were conducted using an optimizing approach that interpolated across the surface using the principle of inverse distance weighting. In the approach, each input point has a local influence that diminishes with the square of distance. All sampling points within a radius of 50 km were used to determine the output value for each location. Obviously, this approach can lead to the loss of information for specific sampling sites. However, we felt that the method allowed us to show a smoothed assessment of surface water Hg distribution across a large area.

The spatial distribution analysis was followed by linear regression to see if the relationship between Hg_t and CH₃Hg⁺ with DOC described above, was general throughout the study region. Using only summer and early autumn data in the regressions restricted Hg concentrations to base-flow conditions and thus those dominated by background geological, pedological, and topographic conditions.

The abundance of flat, wet areas, indicative of wetlands and DOC generation in small watersheds, was predicted using Geographic Information Systems (GIS) (Johnston et al., 1990). We developed models of surface water Hg_t and CH₃Hg⁺ concentrations based on geographical characteristics more suitable for a large-scale application. To do this, we extracted Digital

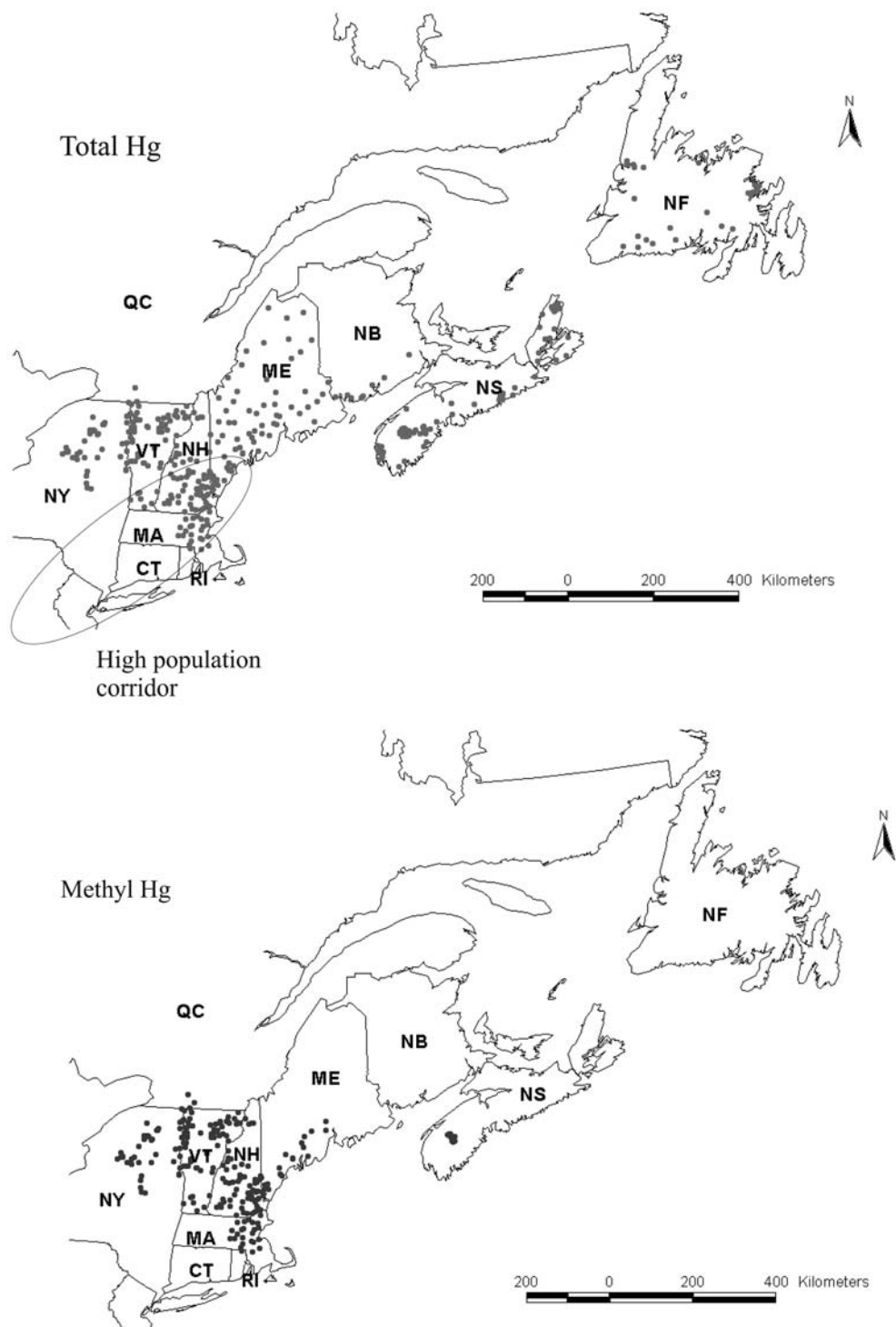


Figure 1. Study region with political boundaries and locations of sites containing Hg_t (top) and CH_3Hg^+ (bottom) data.

Terrain Maps (DTM's) from Natural Resources Canada and United States Geological Survey, National Elevation Dataset maps at 90 m pixel scales. We also used 10m pixel maps for the Adirondack NY region, as these data were available. We identified all catchments where Hg_t and CH_3Hg^+ were collected and analyzed them for area, mean slope, and mean soil drainage index.

The Soil Drainage Index (SDI) provides an assessment of terrain flatness and therefore an indication of potential wetland abundance (MacMillan and Pettapiece 1997). The SDI is calculated as:

$$SDI = 40 * \ln[(A + 1)/(I + 1)^2] \quad (1)$$

where A is calculated flow accumulation within each of the grid cells, and I is the mean slope of the grid cell. SDI values increase as the soil becomes wetter, and thus more likely to support wetlands. We calculated regression equations for each catchment using mean slope, SDI, and catchment area to predict Hg_t and CH_3Hg^+ concentrations in waters draining the catchments for the Northeast region.

Results

Hg_t concentrations ranged widely in surface waters of the region ($n = 831$, Table 1). Values ranged from below the detection level of 0.5 ng l^{-1} to a maximum of 19.5 ng l^{-1} . Mean Hg_t values were greatest in NS, followed by the Adirondacks (NY) and then NF, the sub-region furthest from the HPC. NH and VT, which received very high deposition according to Miller et al. (2005) had the lowest mean water Hg_t concentrations. There were fewer CH_3Hg^+ values ($n = 277$) with none from NB and NF. The highest mean value was again from NS at 0.59 ng l^{-1} , the lowest from VT (0.20 ng l^{-1}). The means for all other sub-regions were near 0.32 ng l^{-1} .

The range in the Hg_t data varied greatly among the different sub-regions (Table 1). VT, NH and ME had lower values, while NY, NS, and NF had the higher (Fig. 2). Hg_t concentrations in NH, VT, and ME are skewed towards the lower concentrations, while the others are more evenly distributed. The distribution of CH_3Hg^+ values follows a pattern somewhat similar to that for Hg_t (Fig. 3),

although we clustered the NH and VT values together as they had similar values and distributions. NS had the highest mean and median values, while NY also showed a pattern towards higher values. NH-VT, ME and MA had lower values. Overall, there was a roughly constant relationship between Hg_t and CH_3Hg^+ (Table 1). When regression was performed on them, we found a significant relationship between the two and an approximate 15:1 $Hg_t:CH_3Hg^+$ ratio during baseflow conditions (Fig. 4).

Despite good relationships across the region, there were a number of sites that did not fit this pattern as the $Hg_t:CH_3Hg^+$ ratios in MA was atypical of the rest of the region (Fig. 4). The remaining sites located in more undeveloped and mostly oligotrophic water bodies all had a strong relationship between Hg_t and CH_3Hg^+ , with the methylated form being roughly 13–15% of Hg_t , depending on location. The MA data had much lower CH_3Hg^+ values than expected. This suggested that the methylation rate was either much lower in this region, or that demethylation was occurring more rapidly at the more urbanized sites (Oremland et al., 1995). It may also be that the MA sites were located in areas with more local anthropogenic influence on water bodies or a lower abundance of wetlands. More work needs to be done to explain this observation.

Two types of spatial patterns seem to occur in northeastern North America (Figs. 5 and 6). Areas within or closest to the HPC were more likely to have higher mean Hg_t values than contiguous regions. There were Hg_t highs to the west of Boston MA and around Portland ME (Fig. 5) and these may be related to industrial sources. There is also an area of unusually high Hg values in northern ME (Fig. 5). Discussions with scientists familiar with local conditions suggest that this intensively farmed area may be affected by historical Hg-based pesticide releases.

We also visually compared the surface water Hg_t and CH_3Hg^+ distribution data to the deposition grid estimated by Miller et al. (this issue). We find a poor correlation between deposition and Hg_t in ME, VT, and NH. Miller et al. predict high deposition levels in mountainous VT and NH due to orographic effects and Hg interception by tree leaves and needles. These are areas where we find low Hg_t and CH_3Hg^+ in surface waters, so it is

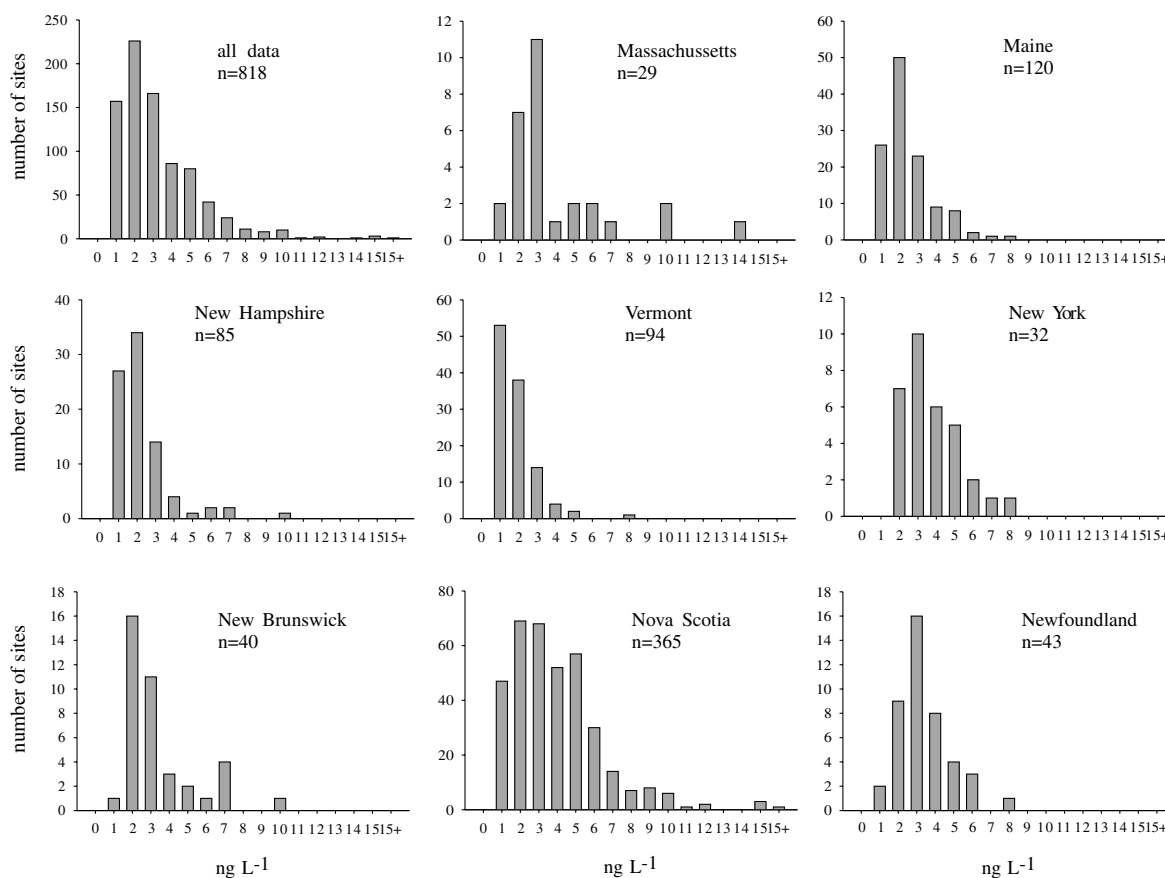


Figure 2. Histogram distribution of Hg_t values in various parts of the study area using only late spring to early fall data.

clear that deposition is not the main driving factor in those regions far from local sources.

The regions furthest from the HPC, northern NY, NS and NF all had areas of high Hg_t concentrations which could not be explained by proximity to anthropogenic sources. Although there may be local effects for Hg_t in MA and ME that could be ascribed to nearby human activities, there clearly are other factors controlling Hg_t concentrations in northern ME, NS and NF. The regional CH_3Hg^+ pattern is similar as that for Hg_t (Fig. 6). There are regional highs in east central and northeast MA–southern NH, as well as in western NS. CH_3Hg^+ was greatest in NS, central MA and southern NH. Unlike for Hg_t , the Boston area did not have higher CH_3Hg^+ values.

Hg_t and CH_3Hg^+ concentrations have been linked to DOC in oligotrophic waters (Lu and Jaffe, 2001) as it has metal complexation capabilities

from natural organic ligands. Our work showed that DOC correlated well with the Hg_t and CH_3Hg^+ concentrations, producing two significant positive correlations (Fig. 7). This suggests that Hg in much of northeastern NA is in large part controlled by the presence of wetlands. $Hg(II)$ and CH_3Hg^+ behave much like many metals in water (Reuter and Perdue 1977; Haitzer et al., 2002). Our data thus suggest that regions with a higher percentage wetland area will have higher surface water Hg concentrations.

Discussion

A number of studies have predicted the abundance of wetlands and therefore the production of DOC in landscapes, using only catchment geographical characteristics. Johnson et al. (1990) used 33

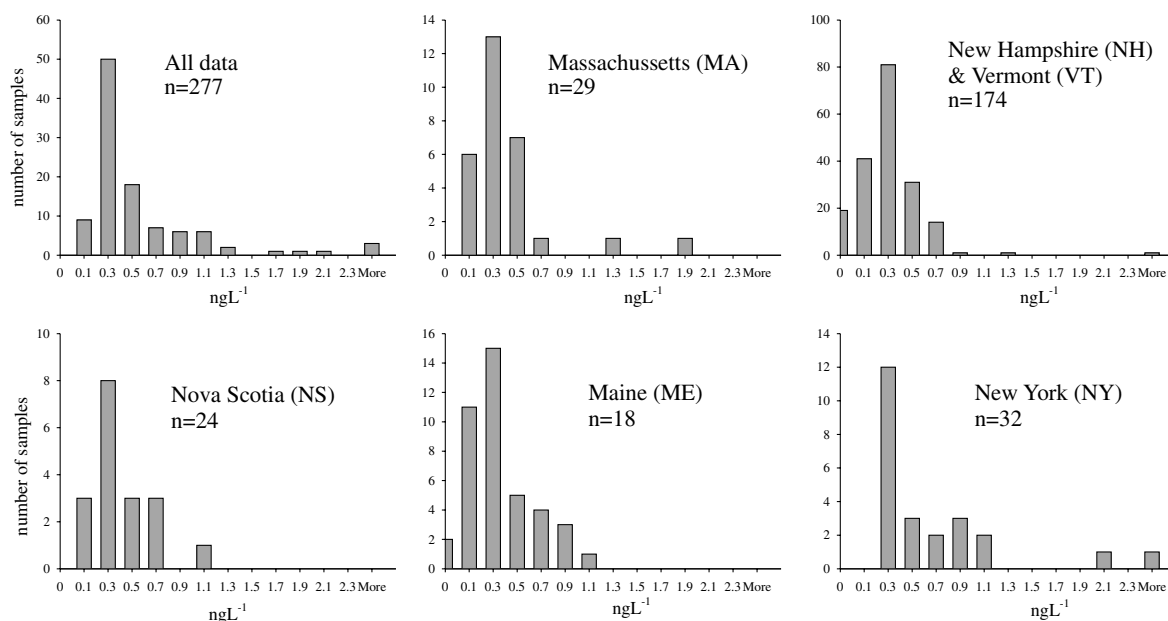


Figure 3. Histogram distribution of MeHg values in various parts of the study area using only late spring to early fall data.

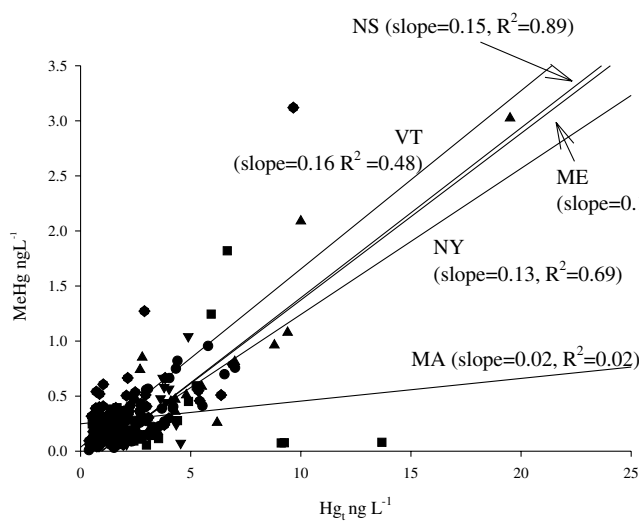


Figure 4. Relationship between Hg_t and CH_3Hg^+ in the samples. Samples at the analytical detection limit were removed from the analysis. Circles are NY, squares-MA, up triangles- NS, down triangles-ME, hexagons-NH, diamonds-VT.

watershed variables in a Geographical Information System (GIS) approach to predict DOC concentrations in streams of Minnesota. Clair and Ehrman (1996, 1998) used a similar approach to predict carbon concentrations and fluxes from watersheds in Atlantic Canada. Using our data, we reasoned that because DOC and Hg are statisti-

cally linked, a simple statistical model using parameters estimated by GIS tools could predict the abundance of wetlands and thus Hg baseflow concentrations.

We developed a simple model using catchment area, mean slope, and the mean drainage index, extractable from DTM's to predict Hg_t and

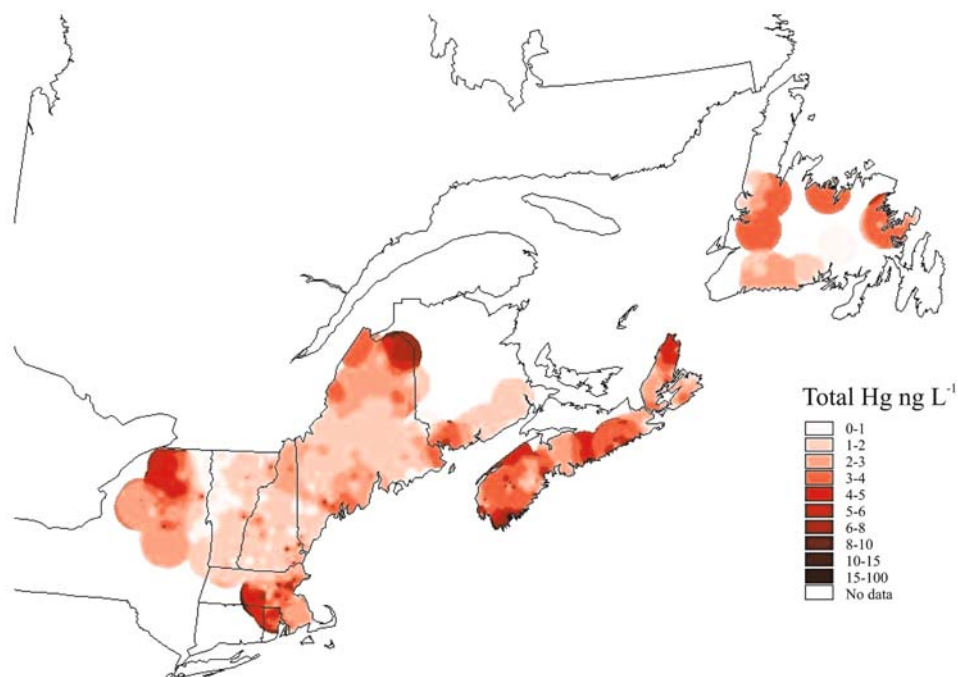


Figure 5. Spatial distribution of late spring – early summer Hg_t data.

CH₃Hg⁺ concentrations in water under baseflow conditions. Using the 90 m DTM's and these three input variables, we were not able to produce a model which could be applied to the entire northeast of NA. We then modified our approach two ways. First, we stratified the data into sub-regional groups, as there were clear differences in topography and concentration among the jurisdictions and then, we used the finer detailed 10 m pixel information for the Adirondack region.

We found more acceptable results using the sub-regional stratification at 90 pixels (Table 2). We calculated predictive Hg_t models with significant or marginally significant *p* values for the Maritime Provinces (NB–NS) and the New England coastal data. The 10m pixel Adirondack DTM allowed us to produce the best predictive model, with a highly significant correlation (*r*²) of 0.49. We were only able to construct a statistically significant CH₃Hg⁺ model with the Adirondack sub-regional data (Table 2). Nevertheless, the overall message of the analysis showed that the poorer the drainage, and the shallower the mean slope, the greater the baseflow Hg_t and CH₃Hg⁺ concentrations.

The success of our approach was inconsistent, in that models could be built for some of the sub-regions but not for others. The data showed two likely reasons for this result. First, for NB, NS, NY and MA (Table 1, Fig. 2) there was enough variation in the input data for a robust model to be built. By contrast, ME, NH, and VT had very narrow Hg concentration and slope data ranges and there was insufficient data spread to allow a statistical model to be constructed. The narrow range in the data, as well as the generally low values are consistent with the fact that the mean slopes are steepest in these three areas and thus are least likely to support wetlands which produce TOC and thus enhance Hg availability.

Secondly, the quality of the DTM data relative to the sizes of the catchments studied was a factor. The best relationship was calculated for the Adirondack sub-region (NY) using 10 m pixel size data as the catchment delineations calculations were more accurate at this resolution. We were also able to compare calculated delineations to those already estimated by ground truthing and found a number of instances where the calculated values were significantly different from the

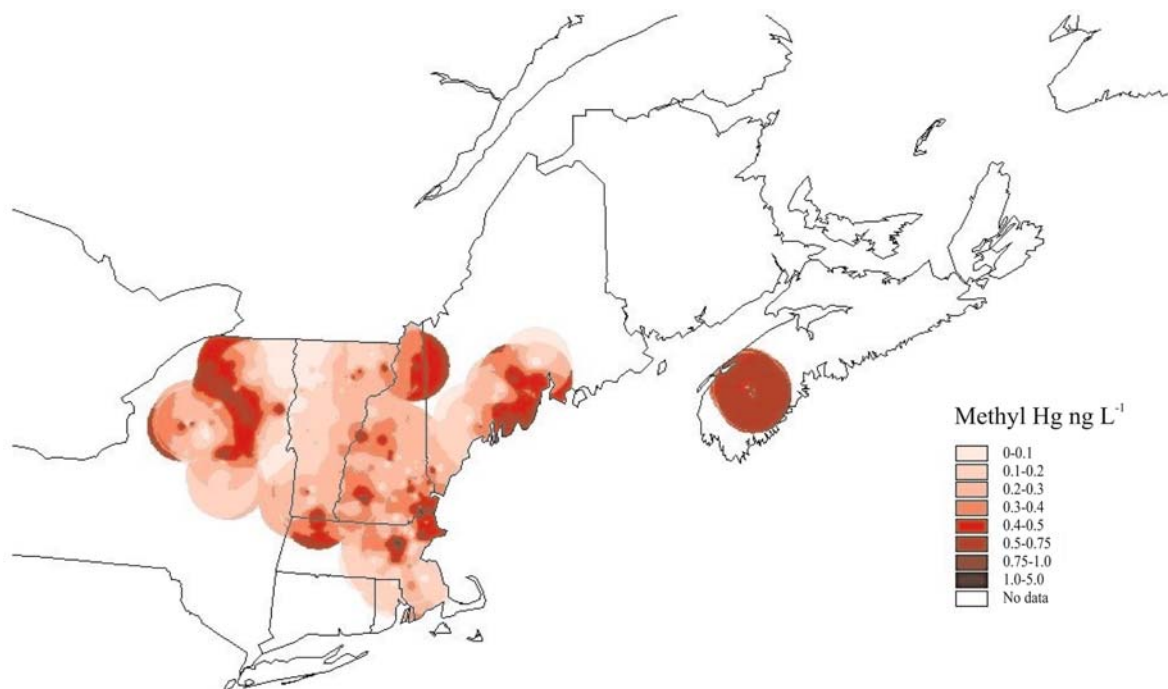


Figure 6. Spatial distribution of late spring – early summer CH_3Hg^+ data.

manually determined ones. In these cases, we used the better local information when it was available. This approach was obviously more accurate and successful, but required much more effort and information than was available for the other regions.

Based on our work, we believe that simple statistical models can be developed to produce estimates of baseflow Hg_t concentrations in areas with a wide range of catchment slopes. For smaller catchments, very high quality DTM and vegetation data are necessary, but these are not yet commonly available for much of North America. The GIS modeling approach was not as successful for CH_3Hg^+ for a number of reasons. First, the problems identified with the Hg_t modeling attempts also apply. Secondly, there were fewer data available for the analysis, so that it was more difficult to reduce the statistical error in the analyses. Finally, the methylation and concurrent demethylation of Hg in soils and wetlands, which we were not able to factor in, added another, unknown level of variability.

Our results show that for much of the region, topography most likely controls the concentration

of Hg in surface waters. In the northeast of North America, there are distinct “hot spots” that can be related to populated areas and are independent of terrain characteristics. Because of two probable mechanisms controlling surface water Hg concentrations (terrain and local sources), it will be difficult to generate GIS models predicting surface water Hg concentrations that can be used with any substantial accuracy.

Despite our inability to produce regionally consistent models predicting the concentration of Hg in surface waters of the northeast of North America, we have used a data set assembled by a wide range of organizations to produce a large-scale picture of the distribution of Hg in the region’s surface waters. These data show that not all parts of northeastern North America are equally susceptible to contamination. Additional research is clearly warranted to explain high Hg in urban water bodies, especially around Boston. There are also relatively undisturbed and remote areas, where long-range transport, combined with catchment characteristics are producing higher aqueous Hg concentration values than would be expected from atmospheric deposition only.

Table 2. Multilinear regression inputs (slope, soil drainage index (SDI), and surface area of catchments as calculated from low resolution digital terrain maps (DTM's)) and regression correlation coefficients for Hg_t and CH_3Hg^+

Region	Area (km ²)	Slope (m km ⁻¹)	SDI	r^2		p	
				Hg_t	Hg_t	CH_3Hg^+	CH_3Hg^+
NB-NS	734 ± 610	2.53 ± 1.6	19.1 ± 8.8	0.33	<0.02	ND	ND
ME	655 ± 1346	3.6 ± 2.6	25.7 ± 10.8	0.19	0.32	ND	ND
VT-NH	248.8 ± 666	4.8 ± 2.1	17.3 ± 8.0	0.15	0.27	0.02	0.9
L. Champlain	2356 ± 1509	5.1 ± 1.4	21.0 ± 26.7	0.26	0.17	0.24	0.19
NE coastal	147 ± 227	1.8 ± 0.8	27.8 ± 8.8	0.34	0.07	0.15	0.36
Adirondacks	54.9 ± 99.7	6.9 ± 3.2	9.7 ± 4.8	0.49	<0.001	0.38	0.008

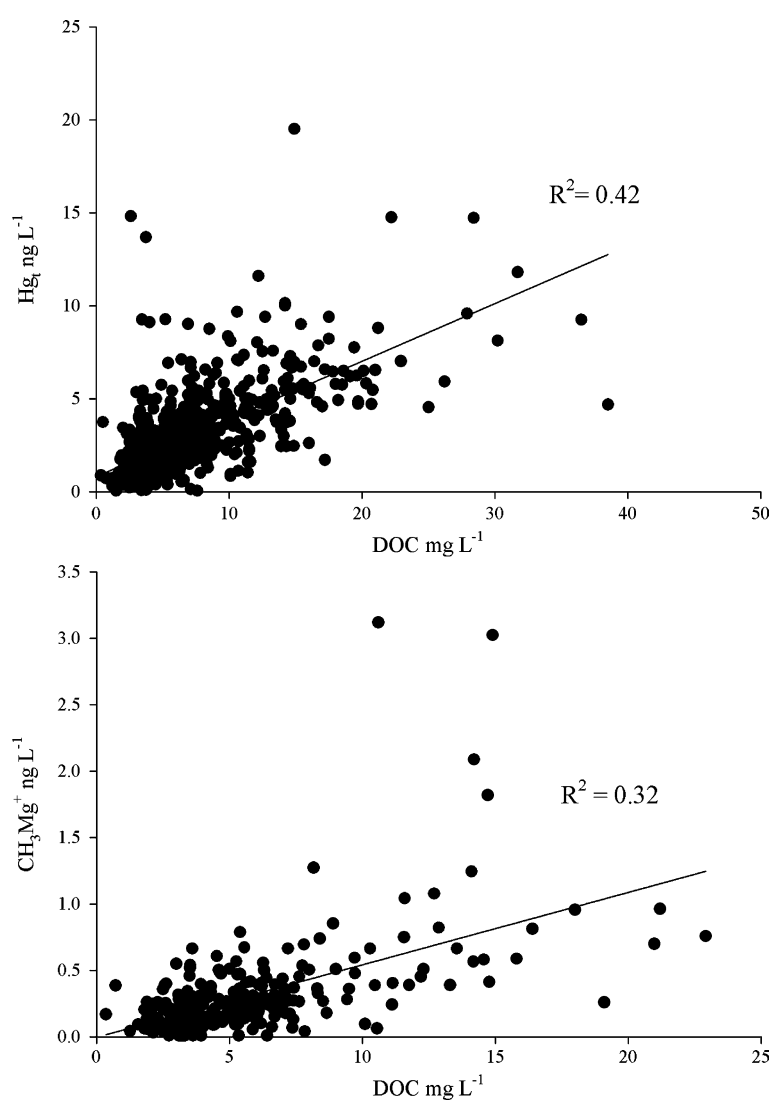


Figure 7. Linear regression between DOC and Hg_t (top) as well as DOC and CH_3Hg^+ (bottom).

Acknowledgements

We thank Wing Goodale of the Biodiversity Research Institute for data management. Dr Fan Rui Meng of the University of New Brunswick assisted us in providing software that we used to calculate catchment sizes. John Dalziel of Fisheries and Oceans Canada provided us with important data from New Brunswick and Nova Scotia.

References

- Briloon. (2004). Biodiversity Research Institute NERC Mercury Project Web site <http://www.briloon.org/NERC/nerc2/index.htm>.
- Clair, T.A. and Ehrman, J.M. (1996). Variation in discharge, dissolved organic carbon and nitrogen export from terrestrial basins with changes in climate: a neural network approach. *Limnol. Oceanogr.* **41**, 921–27.
- Clair, T.A. and Ehrman, J.M. (1998). The influence of changing seasonal climates in modifying discharge, dissolved organic carbon and nitrogen export in wetland influenced rivers: a neural network approach. *Water Resour. Res.* **34**, 447–55.
- Driscoll, C.T., Blette, V., Yan, C., Schofield, C.L., Munson, R. and Holsapple, J. (1995). The role of dissolved organic carbon in the chemistry and bioavailability of mercury in remote Adirondack lakes. *Water Air Soil Pollut.* **80**, 499–508.
- Haitzer, M., Aiken, G.R. and Ryan, J.N. (2002). Binding of mercury (II) to dissolved organic matter: The role of the mercury-to-DOM concentration ratio. *Environ. Sci. Technol.* **36**, 3564–70.
- Johnston, C.A., Detenbeck, N.E. and Niemi, G.J. (1990). The cumulative effect of wetlands on stream water quality and quantity. A landscape approach. *Biogeochemistry* **10**, 105–41.
- Krabbenhoft, D.P., Olson, M.L., Dewild, J.F., Clow, D.W., Striegl, R.G., Dornblaser, M.M. and VanMetre, P. (2002). Mercury Loading and Methylmercury Production and Cycling in High-Altitude Lakes from the Western United States. *Water Air Soil Pollut. Focus.* **2**, 233–49.
- Landis, M.S., Vette, A.F. and Keeler, G.J. (2002). Atmospheric mercury in the Lake Michigan basin: Influence of the Chicago/Gary urban area. *Environ. Sci. Technol.* **36**, 4508–17.
- Lu, X.Q. and Jaffe, R. (2001). Interaction between Hg(II) and natural dissolved organic matter: A fluorescence spectroscopy based study. *Water Res.* **35**, 1793–803.
- MacMillan, R.A. and Pettapiece, W.W. (1997). Soil landscape models: automated landform characterization and generation of soil-landscape models. Tech. Bull. No. 1997-E. Research Branch, Agriculture and Agri-Food Canada, Lethbridge, AB, 75 pp.
- Miller, E., Van Arsdale, A., Keeler, G.J., Chalmers, A., Poissant, L., Kamman, N. and Brulotte, R. (2005). Estimation and mapping of mercury deposition to northeastern North America. *Ecotoxicology* **14**, 53–70.
- Oremland, R.S., Miller, L.G., Dowdle, P., Connell, T. and Barkay, T. (1995). Methylmercury oxidative degradation potentials in contaminated and pristine sediments of the Carson River, Nevada. *Appl. Environ. Microbiol.* **61**, 2745–53.
- Reuter, J.H. and Perdue, E.M. (1977). Importance of heavy metal-organic matter interactions in natural waters. *Geochim. Cosmochim. Acta* **41**, 325–34.
- Rice, K.C. (1999). Trace-element concentrations in streambed sediment across the conterminous United States. *Environ. Sci. Technol.* **33**, 2499–504.
- Shanley, J.B., Schuster, P.F., Reddy, M.M., Roth, D.A., Taylor, H.E. and Aiken, G.R. (2002). Mercury on the move in Vermont. *Eos* **83**, 45–8.
- Shanley, J.B., Kamman, N.C., Clair, T.A. and Evers, D. Physicochemical correlates and inferred Hg delivery mechanisms in watersheds of northeastern North America. *Ecotoxicology* **14**, 125–134.