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Spatial gradients of methylmercury for breeding common loons in the Laurentian Great Lakes region

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Abstract Much of the Laurentian Great Lakes region is a mercury-sensitive landscape, in which atmospheric deposition and waterborne sources of mercury (Hg) have led to high concentrations of bioavailable methylmercury (MeHg) in predatory fish and piscivorous wildlife. Efforts since the early 1990s have established the common loon (*Gavia immer*) as the primary avian indicator for evaluating the

exposure and effects of MeHg in North America. A regional Hg dataset was compiled from multiple loon tissue types and yellow perch (*Perca flavescens*), a preferred prey fish species for loons. Hg exposure in loons and perch was modeled to develop male and female loon units (MLU and FLU, respectively), standardized metrics that represent the estimated blood Hg exposure of a male or female loon for a given loon territory or water body. Using this common endpoint approach to assess loon Hg exposure, the authors demonstrate spatial trends in biotic Hg concentrations, examine MeHg availability in aquatic ecosystems of the Great Lakes region in relation to landscape-level characteristics, and identify areas with potentially significant adverse reproductive impacts to loons and other avian piscivores. Based on 8,101 MLUs, seven biological Hg hotspots were identified in the Great Lakes region. Policy-relevant applications are presented.

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Introduction

Mercury in the Great Lakes region

Mercury (Hg) pollution poses a significant risk to the health of humans and wildlife within the Great Lakes region. Atmospheric deposition is considered the primary source of Hg accumulating in watersheds and waters in the Great Lakes region (Mason and Sullivan 1997; Rolfhus et al. 2003; Wiener et al. 2006). Analyses of sediment cores from this and other geographic regions show that most of this Hg is derived from anthropogenic sources (Swain et al. 1992; Engstrom and Swain 1997; Lockhart et al. 1998;

Lorey and Driscoll 1999; Lamborg et al. 2002), primarily fossil fuel (coal) combustion (Pacyna et al. 2006). In addition, a growing body of evidence indicates that atmospheric deposition is the primary source of Hg accumulating as methylmercury (MeHg) in aquatic food webs and freshwater fish (Fitzgerald et al. 1998; Hammerschmidt and Fitzgerald 2005, 2006; Wiener et al. 2006; Harris et al. 2007; Munthe et al. 2007). MeHg is produced from inorganic Hg by aquatic sulfate-reducing bacteria (Compeau and Bartha 1985; Gilmour et al. 1992), which serve to integrate the environmental cycling of sulfur and Hg. Wetlands are a primary location for Hg methylation (Krabbenhoft et al. 1995), and aquatic food webs are the primary pathway for exposure of humans and wildlife to MeHg (NRC 2000; Mergler et al. 2007; Scheuhammer et al. 2007). Concentrations of MeHg in fish from many lakes in the Great Lakes region exceed criteria for the protection of human health (USEPA 2009), and much of the region is a Hg-sensitive landscape in which atmospheric deposition has led to high concentrations of MeHg in predatory fish and piscivorous wildlife. Concern for adverse ecosystem impacts has recently grown based on new evidence that the bioavailability of MeHg in the Great Lakes region is increasing, based on game fish in Minnesota (Monson 2009) and Lake Erie (Bhavsar et al. 2010) and loons in Wisconsin (Meyer et al. 2011).

Mercury in common loons

Efforts since the early 1990s have established the common loon (*Gavia immer*) as the primary avian indicator for evaluating the exposure and effects of MeHg availability in freshwater lakes of North America (Evers 2006; Evers et al. 2008, 2010). The common loon's elevated stature is based on well-defined continental-scale exposure profiles for Hg (Evers et al. 1998, 2003; Scheuhammer et al. 2001) that have helped identify biological Hg hotspots in northeastern North America (Evers et al. 2007), and established lowest observed adverse effect levels (Barr 1986; Nocera and Taylor 1998; Burgess et al. 1998; Evers et al. 2003, 2004, 2008; Burgess and Meyer 2008) and risk assessment models (Nacci et al. 2005). Loon studies have also contributed to the scientific community's growing knowledge of Hg pharmacokinetics (Fournier et al. 2002; Kenow et al. 2003, 2011) and of confounding factors such as prey availability (Merrill et al. 2005). Instead of individual-based laboratory studies that were formerly used to extrapolate potential avian effects (Heinz 1979), recent risk assessment efforts with the common loon combine laboratory (Fournier et al. 2002; Kenow et al. 2003, 2011) and field studies (Burgess et al. 2005; Burgess and Meyer 2008; Evers et al. 2008) with developing population models (Nacci et al. 2005; Evers 2007; Gear et al. 2009; Evers et al. 2010). This body of

knowledge is the basis for the common loon as a principal indicator of MeHg availability for freshwater avian piscivores for (1) developing wildlife criterion values of interest to states (Evers et al. 2004) and (2) inclusion within national Hg monitoring programs in Canada (Morrison 2011) and in the United States (Mason et al. 2005; Wolfe et al. 2007).

MeHg, one of the most bioavailable forms of Hg, is highly neurotoxic to both the adult and developing brain (Clarkson and Magos 2006). In fish, birds, and mammals MeHg in reproducing females readily passes to the developing egg or embryo (Evers et al. 2003; Hammerschmidt and Sandheinrich 2005; Heinz et al. 2010), life stages that are much more sensitive than the adult to acute MeHg exposure (Wiener et al. 2003; Evers et al. 2003; Scheuhammer et al. 2007). It is often difficult to determine the severity of adverse effects of specific contaminants to wildlife, particularly in a non-laboratory setting. However, common loons are a primary study organism for Hg contamination. Mercury is associated with adverse effects at sub-lethal exposure levels (Evers et al. 2008; Burgess and Meyer 2008), and a LOAEL (lowest observed adverse effect level) can serve as a simple and convenient benchmark for assessing potential injury to wildlife due to Hg contamination. Effects of Hg at 3.0 $\mu\text{g/g}$, wet weight (ww), in adult common loon blood include aberrant incubation behavior, lethargy, (Evers et al. 2008), higher average corticosterone levels, and increased developmental instability as indicated by flight feather asymmetry (Evers et al. 2004). Although adult survival does not appear to be impaired to a detectable level (Mitro et al. 2008), adult Hg exposure does appear to be highly correlated with reproductive impacts, including significantly lower fledging success once exceeding 3.0 $\mu\text{g/g}$, ww (e.g., >41% fewer fledged young based on Evers et al. (2008) and Burgess and Meyer (2008)). Diminished reproductive success, in particular, can have adverse population-level consequences for fish and wildlife. Using behavioral and reproductive endpoints, loons with blood Hg concentrations below 1.0 $\mu\text{g/g}$ are considered to be at little to no risk for adverse effects, therefore establishing a NOAEL ("no observed adverse effect level," Evers et al. 2004). While loons with 1.0–3.0 $\mu\text{g/g}$ Hg in blood, or the equivalent in other tissues, have demonstrated adverse effects such as physiological changes, aberrant behavior, and reproductive deficiencies (Evers et al. 2008; Burgess and Meyer 2008), full understanding of these effects remains undefined. Although 3.0 $\mu\text{g/g}$ adult blood Hg has been widely recognized as the LOAEL for the species (Evers et al. 2007, 2008), loons with levels above 4.0 $\mu\text{g/g}$ have been categorized at an extra-high risk that is related to observed behavioral changes in the field, such as abnormal and diminished incubation times (Evers et al. 2004), and to the 50%

reduction in productivity associated with this Hg exposure level (Burgess and Meyer 2008).

The need to develop a common unit for loon and perch Hg data concentrations

To best evaluate and utilize existing data from various biotic compartments, a single common unit for Hg concentrations is preferred. Since common loon Hg data are from multiple tissues, sexes, and ages including adult male and female blood, juvenile blood, and loon eggs, comparisons between locations and years can be difficult to conduct or assess. Moreover, loon Hg data are sparse for some regions and collection is quite costly and time-consuming, as it often involves intensive field capture operations. Yellow perch (*Perca flavescens*) are a primary prey species of common loons and the preferred prey size, depending upon the size of the loon, varies from 5 to 25 cm in length (Barr 1996). Perch appear to be a common prey species for loons across their range (Evers et al. 2004; Burgess and Hobson 2006; Burgess and Meyer 2008). As Hg exposure in loons is based nearly exclusively on diet, perch Hg concentrations may be used to predict loon Hg concentrations. However, all data must be directly comparable, by relating samples taken at the same times and locations to create reliable regression models between tissue types. A common unit that incorporates all data types makes it feasible to undertake a large-scale spatial analysis of loon Hg exposure, and explore possible population-level effects.

The objectives of this study are to (1) develop linkages between Hg in fish and Hg in the blood and eggs of the common loon, and thus provide a common endpoint for projected MeHg availability in piscivorous wildlife; (2) determine Hg exposure in loons in the Great Lakes region, (3) demonstrate spatial trends in biotic Hg concentrations, and (4) identify potential landscape-level factors influencing wildlife Hg levels in the Great Lakes region. This paper is an enhanced geographic extension of Evers et al. (2007), and the results presented are directly comparable to northeastern United States and eastern Canada. However, the use of a single standardized unit to generate Hg concentrations for the Great Lakes region and the additional spatial analysis components presented here are unique to this paper. The first phase of this investigation involves normalizing data from different individuals into standard “loon tissue” units to assess the risk of Hg exposure to piscivorous birds. A second phase involves the use of yellow perch regression models to model loon Hg exposure based on perch exposure. The third phase of analysis involves application of effects models to the datasets created in the first two phases, in order to create a spatial model for potential impacts of Hg exposure to common loons in the Great Lakes region, with dual goals of

identifying potential piscivorous wildlife Hg hotspots and elucidating the environmental variables associated with loon Hg exposure.

Methods

Study area

The Great Lakes region, as defined for this study, encompasses the Great Lakes themselves, Lake Champlain, the entirety of the states of Illinois, Indiana, Michigan, Minnesota, New York (with the exception of Long Island), Ohio, Pennsylvania, and Wisconsin, and the province of Ontario. Information on the hydrology, geology, and land use of this region may be found in Wiener et al. (2011). For spatial modeling in this study, data from the Great Lakes and Lake Champlain proper were excluded, as loons generally do not breed on the Great Lakes themselves (a recognized exception is Isle Royale, Michigan; Evers et al. 2010).

Data sources and loon unit modeling

Loon and perch data sources

Loon data were from long-term datasets held by the Biodiversity Research Institute, the Wisconsin Department of Natural Resources, and Environment Canada's National Wildlife Research Centre. Yellow perch data were part of a large fish dataset from state, federal, and other sources in the Great Lakes region, developed as part of the Great Lakes Mercury Project (details on this dataset may be found in Monson et al. 2011). Loon samples were analyzed as in Evers et al. (1998), Evers et al. (2003, 2008), and Meyer et al. (2011). Only data between the years of 1990 and 2009 were included, and only perch data from fish 5–25 cm in length, which are common prey items for loons (Barr 1996). Chicks of unknown body weight and chicks greater than 6 weeks of age were excluded. Data of questionable quality were eliminated from the database, such as records lacking location information or dates, or with Hg values below laboratory detection limits of 0.025 µg/g. Analysis of samples for total mercury (THg) occurred at several laboratories, and the authors acknowledge the possibility for inter-laboratory error in this study. However, the level of analysis error between laboratories is relatively minor in comparison to the spatial and temporal signal presented here and in companion articles (Monson et al. 2011; Meyer et al. 2011).

With the resulting dataset, regression models developed in this study and elsewhere (Evers et al. 2003; 2004; Kamman et al. 2004; Evers et al. 2004) were used to convert common loon egg Hg ($n = 322$), adult and chick

blood Hg ($n = 1,563$) and yellow perch Hg values ($n = 4,371$ fillet and 2,246 whole body records in original dataset) from the Great Lakes region into common units.

Loon unit modeling

Subsets of the loon data, in which there were multiple Hg data points from a single territory and year, were used to develop relationships between Hg in different tissues. These models were then applied to the larger dataset to present data from all tissue types, territories and years, in a common unit, the so-called “female loon unit” (FLU) to represent adult female loon Hg exposures, and the “male loon unit” (MLU) to predict adult male exposures. Male loons on the breeding grounds tend to have higher Hg than females, presumably due to the depuration of female body Hg into eggs and to differences in diet (males are larger and eat larger prey; Barr 1996). Male blood Hg, juvenile blood Hg from several age classes, and egg Hg were each separately regressed with female blood Hg to convert all tissues to FLUs. Female adult and juvenile blood concentrations were also converted into MLUs. While there is a good published regression model for relating egg Hg to female loon Hg (Evers et al. 2003), no such relationship exists for male loon Hg; as such, presentation of Hg data in FLUs presents a different picture than in MLUs, since FLUs include egg data and are the more universal unit. MLUs, however, represent the often more severe Hg exposures of larger male loons in the region, and are useful for examining the potential for population-level adverse effects of Hg exposure.

JMP Version 9.0 (2010) was used to conduct an initial multivariate analysis of female, male, and chick blood Hg values. Loon blood Hg concentrations, like most contaminant data, are highly right-skewed and are not normally distributed, so blood Hg concentrations were natural log-transformed to normalize the data. The authors regressed blood Hg values from the same territories and years. There is a strong relationship ($r^2 = 0.77$) between chick age and body weight (unpub. data; Online Resource 1) based on a large dataset of known-age juvenile common loons from states and provinces throughout northern North America. This relationship was used to assign an estimated age, in days, to all juveniles of known weight in the dataset. Chicks were split into three age categories: <4, 4–6, and >6 weeks, based on biological differences in Hg exposure and depuration of Hg (Evers et al. 2010; Online Resource 2). Female blood Hg was separately regressed with Hg in the blood of young chicks for the three age categories. Initial analysis indicated that Hg values from older chicks (>6 weeks of age) were not correlated with female, male, or younger chick Hg concentrations, and Hg exposure was not correlated with body weight in chicks for any of the three age groups examined. Adult male and female Hg

exposures were also not well correlated with body weight ($r^2 = 0.02$; Online Resource 2).

Calculation of FLUs and MLUs from adult blood Hg data

The authors regressed female blood Hg with male blood Hg from the same territories and years ($n = 276$; Online Resource 3). There is no clear dependent relationship between the two variables, so an orthogonal regression method with an assumption of equal variances was used to minimize residuals along both X – Y -axes. This method also allows for prediction of both x and y values (Freund et al. 2003). Thirteen cases were excluded from the analysis as being outside the 95% density ellipse, which is also the confidence curve for the regression (e.g., indicating the equivalent of a 95% confidence interval for the data; SAS Institute Inc. 2010). The regression and 95% confidence limits (CLs) between male and female natural log-transformed blood Hg concentrations are presented in Eqs. 1 and 2. ♂ Hg represents male and ♀ Hg female blood Hg concentrations.

FLUs from male blood concentrations:

$$FLU = e^{-0.37798+1.065584 \cdot \ln(MLU)} \quad 95\%CLs: 0.96 - 1.18$$

$$r = 0.75 \quad (1)$$

MLUs from female blood concentrations:

$$MLU = e^{0.354719+0.938452 \cdot \ln(FLU)} \quad 95\%CLs: 0.85 - 1.04$$

$$r = 0.75 \quad (2)$$

The above models well described the observed data ($r = 1.00$, $n = 985$). The full dataset includes the observed blood Hg concentrations for male and female loons, as well as estimated values.

Calculation of FLUs and MLUs from juvenile blood Hg data

Within about three weeks of hatching, loon chick blood Hg concentrations become independent of the original maternal transfer of MeHg into the egg (Kenow et al. 2003). Orthogonal regression with an assumption of equal variances was used to model chick blood Hg concentrations with adult female blood concentrations (Eqs. 3–4) and male blood concentrations (Eqs. 5–6) from the same territories and years (Online Resource 3). As mentioned previously, blood Hg from chicks greater than 6 weeks of age was not significantly correlated with adult blood Hg or younger chick Hg, and thus was not modeled. For territories where there were Hg values for an adult and both chicks, and the chicks fell into the same age group as estimated by body weight, the chick Hg values were averaged prior to regression.

Both female and chick Hg concentrations were natural log transformed and regressed ($n = 80$ complete cases for <4 week old chicks; $n = 127$ for 4–6 week chicks). Seven and six outliers were excluded from the <4 week and 4–6 week old regression models, respectively (Eqs. 3–4). For the regression model including male blood and chick blood Hg, six outliers were excluded from analysis for young chicks (leaving $n = 89$) and nine for 4–6 week old chicks ($n = 176$; Eqs. 5–6). Outliers are likely due largely to differences in the growth of compromised chicks, as sibling rivalry is an important mechanism for population regulation (Evers et al. 2010).

FLUs from <4 week old chick blood concentrations

$$FLU = e^{1.102286 + 0.464995 \cdot \ln(\text{JuvHg})} \quad 95\%CLs: 0.34 - 0.60$$

$$r = 0.64 \quad (3)$$

FLUs from 4 to 6 week old chick blood concentrations

$$FLU = e^{1.552003 + 0.674632 \cdot \ln(\text{JuvHg})} \quad 95\%CLs: 0.55 - 0.81$$

$$r = 0.68 \quad (4)$$

MLUs from <4 week old chick blood concentrations

$$MLU = e^{1.820043 + 0.648913 \cdot \ln(\text{JuvHg})} \quad 95\%CLs: 0.50 - 0.82$$

$$r = 0.66 \quad (5)$$

MLUs from 4 to 6 week old chick blood concentrations

$$MLU = e^{1.68781 + 0.587552 \cdot \ln(\text{JuvHg})} \quad 95\%CLs: 0.48 - 0.71$$

$$r = 0.61 \quad (6)$$

Calculation of FLUs from egg Hg data

Fresh wet weight egg Hg concentrations were highly related to female concentrations ($FLU = 0.2238 + 1.5544 \cdot \text{EggHg}$, $r^2 = 0.79$) from the same territories based on the relationship established in Evers et al. (2003), using 108 records with paired hen blood and egg Hg concentrations (1988–2001; Online Resource 3). Samples were from eight states in the US, including New York, Minnesota, Michigan, and Wisconsin.

Calculation of FLUs and MLUs from yellow perch data

A 1998–2003 yellow perch and common loon dataset from western Maine was used to develop loon-perch regression models, as all perch and loon data were collected together from the same lakes during the same time periods ($n = 845$ territory-years, with 95% of loon and perch samples collected less than a week apart; Evers et al. 2004; Online Resource 4). While no standardized study of loon prey preferences and availability has occurred across their range, existing studies support Barr's (1996) findings of yellow perch as a common prey for loons (Evers et al. 2004; Burgess and Hobson 2006). Preferred loon prey size

varies to some extent with loon body size, as shown in Barr 1996; the fish size classes used in Barr's work were used to develop regression models between perch Hg and adult male and female loon Hg in this study's dataset (Eqs. 7–14), where YPHg represents a yellow perch whole body Hg value ($\mu\text{g/g}$ wet weight). Fillet perch samples were converted to whole-body Hg equivalents after Evers et al. (2007): fillet Hg = $[1.63 \cdot \text{whole body Hg}] + 0.06$ (Kamman et al. 2004).

Loon Units from small fish (5–10 cm)

$$FLU = 19.34 \cdot \text{YPHg} + 0.453 \quad R^2 = 0.79 \quad (7)$$

$$MLU = 24.959 \cdot \text{YPHg} + 0.93 \quad R^2 = 0.54 \quad (8)$$

Loon Units from medium-sized fish (10–15 cm)

$$FLU = 10.644 \cdot \text{YPHg} + 0.5079 \quad R^2 = 0.51 \quad (9)$$

$$MLU = 15.17 \cdot \text{YPHg} + 0.75 \quad R^2 = 0.47 \quad (10)$$

Loon Units from large fish (15–20 cm)

$$FLU = 9.2707 \cdot \text{YPHg} + 0.2884 \quad R^2 = 0.74 \quad (11)$$

$$MLU = 12.912 \cdot \text{YPHg} + 0.534 \quad R^2 = 0.59 \quad (12)$$

Loon Units from extra-large fish (20–25 cm)

$$FLU = 5.4695 \cdot \text{YPHg} + 0.4617 \quad R^2 = 0.90 \quad (13)$$

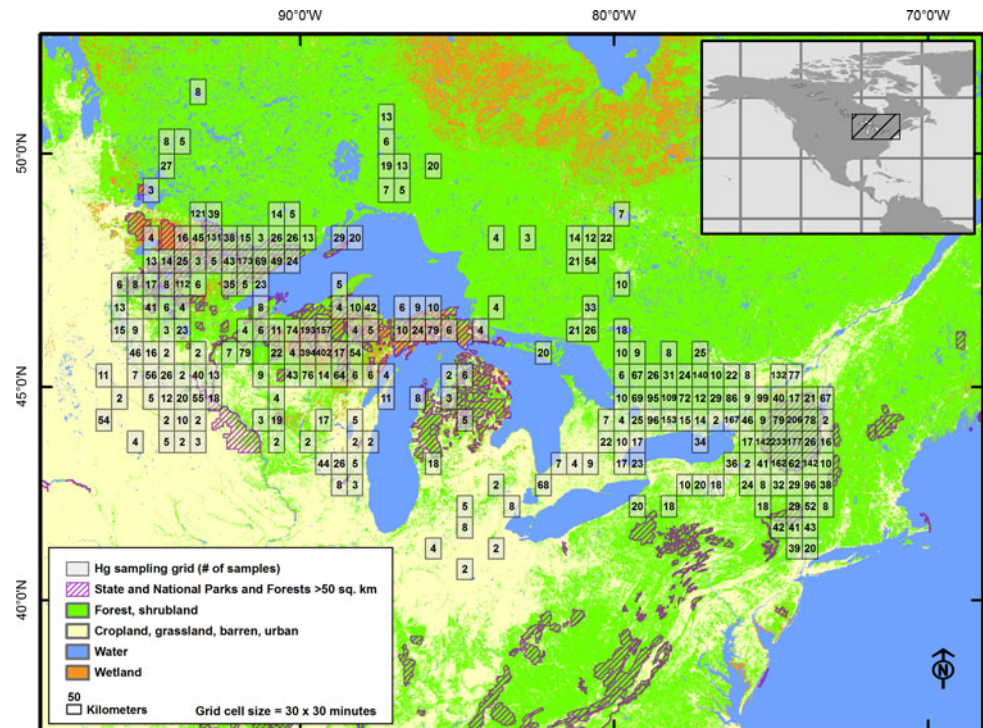
$$MLU = 7.5201 \cdot \text{YPHg} + 0.9157 \quad R^2 = 0.71 \quad (14)$$

Spatial analyses

Mapping of perch and loon data

The loon and perch dataset was mapped using a $0.5^\circ \times 0.5^\circ$ polygon grid interval with GIS (geographic information system) techniques, to summarize the data and target locations with sufficient data to characterize Hg exposures in wildlife (Fig. 1). Gridding of data was modeled on the process developed in Evers et al. 2007. In this case, the grid was offset by 0.000001° to prevent data with imprecise geographic attributions (to the nearest half degree, for instance) from falling exactly on the grid lines between cells. The authors conducted a power analysis to determine the number of samples per cell required to be able to detect biological threshold limits. Given $\alpha = 0.05$ and a relative error rate of 0.86 (based on the proportion of the mean FLU value for the entire dataset represented by a single standard deviation from the mean), the estimated minimum sample size per grid cell necessary to adequately characterize Hg exposure in wildlife was $n = 7$ for FLU values and $n = 9$ for MLU values. The resulting spatial patterns were consistent between FLUs and MLUs, but male loon units illustrated a broader range of exposure concentrations, including higher concentrations than those often exhibited by female loons. As such, data are presented in the form of

Fig. 1 Distribution of loon and perch Hg observations across the Laurentian Great Lakes region ($n = 8,101$ data points), from which male loon units (MLUs) were calculated. Sample size is noted per grid cell. State and federally owned public lands are noted in purple



MLUs, which can provide more detailed spatial patterns. Locations with major emissions sources (e.g. chemical plants, coal-fired power plants, and other sources that emitted greater than 45 kg of Hg per year) were identified using 2002 Hg emissions data (Fig. 2; USEPA 2002) and 2002 Hg data extracted from Canada's National Pollution Release Inventory 1994–2008 (Fig. 2; Environment Canada 2010). USEPA-designated Superfund sites with Hg contamination were identified from the National Priorities List (USEPA 2010) and obtained from TOXMAP (US National Library of Medicine 2010).

Geospatial analyses and kriging along watershed group boundaries

In four focal areas for which there were large amounts of data available, ordinary kriging (a more sophisticated geospatial analysis of wildlife Hg exposure) was conducted using ArcInfo v.10 Geostatistical Analyst (ESRI 2010). The MLU Hg spatial data were fitted to Gaussian models and the data checked against these models for best fit by inspecting the semi-variogram generated by the Geostatistical Wizard. The data optimization routine developed in ArcGIS was used for all regions except north-central Wisconsin, where this procedure resulted in poor model fit; in this case lag size and number of lags was varied to optimize model fit and reduce mean standard error as estimated by cross validation procedures. Groups of watersheds containing each target area were used to create

point boundaries for kriging. Kriged areas were further limited to regions with higher densities of data points within these target watersheds, because the sparse data resulted in very low confidence prediction surfaces. Four areas were chosen for further examination with these more advanced statistical spatial analyses, based upon potential hotspots identified in the grid cell map (Fig. 2), density of available data, and accessibility to previously published studies. Water bodies and agricultural ecumene were added to several figures to aid in interpretation (National Atlas of the United States, US Geological Survey, and Statistics Canada 2004).

Results

For lakes with elevated MeHg availability, it has been shown that on average, large loons have higher blood Hg concentrations than smaller loons (e.g., larger males have significantly greater blood and feather Hg concentrations than females; Evers et al. 1998, 2004). Using a test case of Rainy Lake in Minnesota, the water body with the greatest number of adult blood Hg values (with body weights) in this study ($n = 53$), it is found that when both sexes are considered together, there is indeed a relationship between body mass and blood Hg ($R^2 = 0.24$), but that when the sexes were examined individually, as for modeling efforts in this study, this relationship disappears. Similar patterns

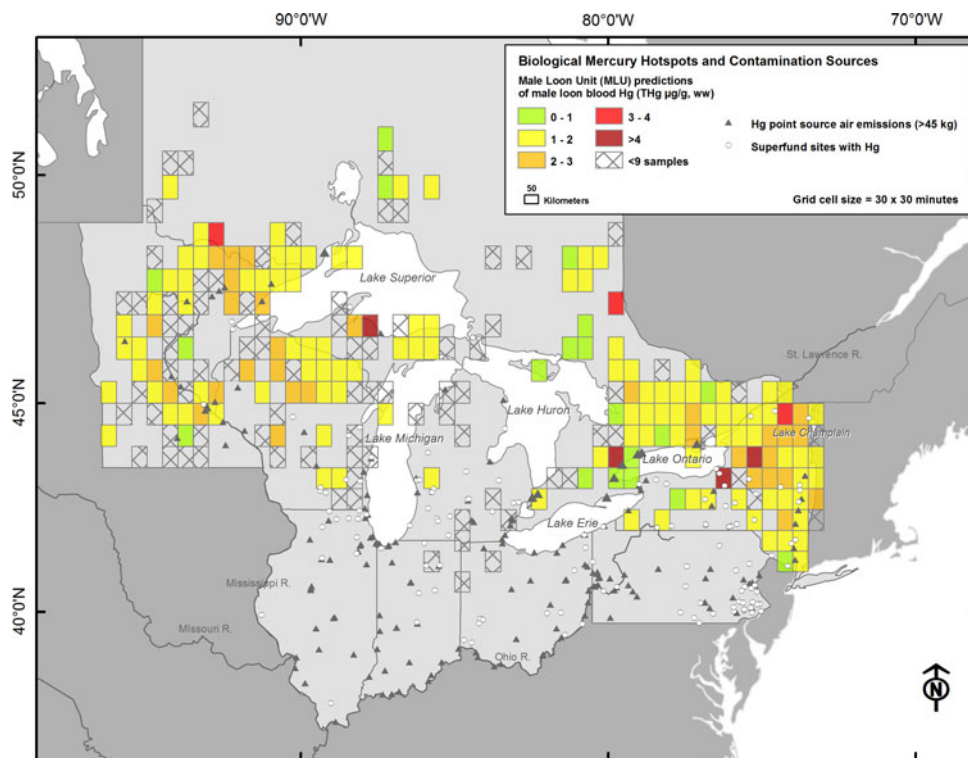


Fig. 2 Distribution of biological Hg hotspots and major Hg emissions sources in the Great Lakes region for 30 × 30 min grid cells with ≥9 samples per grid cell (based on common loon and yellow perch data; $n = 8,101$). Grid cells with fewer than 9 samples were excluded from analysis. Values are male loon units (MLUs), total Hg concentrations in male loon blood that are actual or estimated from loon tissues and yellow perch tissues ($\mu\text{g/g}$, wet weight). Locations with major air emissions sources (e.g. chemical plants, coal-fired

power plants, and other sources that emitted greater than 45 kg of Hg per year) were identified using the USEPA's 2002 Hg emissions data (USEPA 2002) and 2002 Hg data extracted from Canada's National Pollution Release Inventory 1994–2008 (Environment Canada 2010), and are marked on the map with gray triangles. USEPA-designated Superfund sites with Hg contamination (white circles) were identified from the National Priorities List (USEPA 2010) and obtained from TOXMAP (US National Library of Medicine 2010)

were exhibited for Flagstaff Lake in Maine (a large lake for which BRI has 72 adult blood Hg concentrations); male and female blood Hg together was correlated with body weight ($R^2 = 0.23$), but either female or male blood Hg alone was not. Thus, for regional scale modeling and geospatial analysis used in this study, the authors purposely did not account for body weight.

Spatial distribution of Hg concentrations across the Great Lakes region is heterogenous (Fig. 2). Of the 8,423 FLU values used in analyses, 3.7% were above the lowest observed adverse effect level (LOAEL) for loons of 3.0 $\mu\text{g/g}$ wet weight in blood (Table 1). The average FLU value across the Laurentian Great Lakes region (excluding the Great Lakes proper) was 1.21 $\mu\text{g/g}$, ww (± 1.06). Fifty percent of FLU values were above 1.0 $\mu\text{g/g}$. For the 8,101 MLU values in the dataset, a higher proportion than for females was above LOAELs (9.8%): 82% were above 1.0 $\mu\text{g/g}$ and the average MLU concentration across all areas was $1.80 \pm 1.44 \mu\text{g/g}$. Regionwide FLUs and MLUs were significantly different (Wilcoxon signed ranks test, $P \leq 0.0001$).

Four focal regions using MLU concentrations are examined in greater spatial detail for the reasons noted above (Table 2). They include northeastern Minnesota, northern Wisconsin and the Upper Peninsula of Michigan, southern Ontario, and northern New York. The data in each focal area (also defined by sections of watersheds with high data densities) produced geographically continuous predictions of MLUs, based upon data values and geographic proximity to other samples (Figs. 3, 4, 5, 6). With caution, geostatistical analysis of the focal areas serves to further elucidate biological Hg patterns within each location. Figure 3, for instance, shows detailed patterns of predicted biological Hg exposure, expressed as MLUs, in northeastern Minnesota. Three watersheds containing a concentration of data in this area were collectively kriged, with predicted MLUs shown in the main illustration and the prediction standard errors shown in the inset map. In this area of Minnesota, the standard errors indicate little geographic correlation; that is, the proximity of Hg samples is unrelated with similarity in Hg values, and a geospatial analysis as conducted here thus has a high level of

Table 1 Summary statistics for combined loon and perch mercury (Hg) concentrations ($\mu\text{g/g}$, wet weight) in the Laurentian Great Lakes region

Category	Sample size	Hg mean \pm SD ($\mu\text{g/g}$, ww)	Hg range ($\mu\text{g/g}$, ww) [#]	% of data <1 $\mu\text{g/g}$, ww	% of data ≥ 1 and <2 $\mu\text{g/g}$, ww	% of data ≥ 2 and <3 $\mu\text{g/g}$, ww	% of data ≥ 3 and <4 $\mu\text{g/g}$, ww	% of data ≥ 4 $\mu\text{g/g}$, ww
Female loon unit (FLU)	8423	1.2 \pm 1.1 ^a	0.1–41.1	50.4% ($n = 4244$)	38.1% ($n = 3209$)	7.8% ($n = 657$)	2.3% ($n = 195$)	1.4% ($n = 118$)
Male loon unit (MLU)	8101	1.8 \pm 1.4 ^b	0.1–53.3	18.1% ($n = 1462$)	53.5% ($n = 4335$)	18.7% ($n = 1513$)	6.0% ($n = 486$)	3.8% ($n = 305$)

Column means with *different letters* are significantly different (Wilcoxon signed ranks test, $P \leq 0.0001$)

[#] Maximum Hg concentrations for both FLU and MLU (in the value range) are calculated values based on yellow perch Hg concentrations

Table 2 Summary of mercury (Hg) concentrations ($\mu\text{g/g}$, wet weight) in male loon units (MLUs) for four focal areas in the Laurentian Great Lakes region

Focal area	State/province	Focal area (km^2)	Sample size	Hg mean \pm SD ($\mu\text{g/g}$, ww)	Hg range ($\mu\text{g/g}$, ww) [#]	% of data <1 $\mu\text{g/g}$, ww	% of data ≥ 1 and <2 $\mu\text{g/g}$, ww	% of data ≥ 2 and <3 $\mu\text{g/g}$, ww	% of data ≥ 3 and <4 $\mu\text{g/g}$, ww	% of data ≥ 4 $\mu\text{g/g}$, ww
Northern Minnesota	Minnesota	30,676	798	1.9 \pm 1.0 ^a	0.3–9.7	11.4% ($n = 91$)	51.4% ($n = 410$)	25.4% ($n = 203$)	8.0% ($n = 64$)	3.8% ($n = 30$)
Northern Wisconsin and the Upper Peninsula of Michigan	Wisconsin, Michigan	82,562	1687	2.0 \pm 2.0 ^a	0.4–53.3	11.4% ($n = 193$)	54.1% ($n = 912$)	22.4% ($n = 378$)	7.9% ($n = 133$)	4.2% ($n = 71$)
Southern Ontario	Ontario	58,060	1064	1.5 \pm 0.9 ^b	0.1–7.2	30.6% ($n = 326$)	52.4% ($n = 558$)	11.6% ($n = 123$)	3.8% ($n = 40$)	1.6% ($n = 17$)
Northern New York	New York	46,981	1471	2.4 \pm 1.4 ^c	0.3–14.4	6.1% ($n = 89$)	42.1% ($n = 619$)	30.3% ($n = 447$)	12.0% ($n = 177$)	9.4% ($n = 139$)

Column means with *different letters* are significantly different (Wilcoxon pairwise comparison, $\alpha = 0.05$, $P \leq 0.0001$)

[#] Maximum Hg concentrations for all focal areas are calculated values based on yellow perch Hg concentrations

Fig. 3 Predicted male loon blood mercury values ($\mu\text{g/g}$ of total Hg in blood, ww) for a focal area in northeastern Minnesota, based on common loon and yellow perch Hg data. Inset map shows standard errors of the model-based predictions across the region

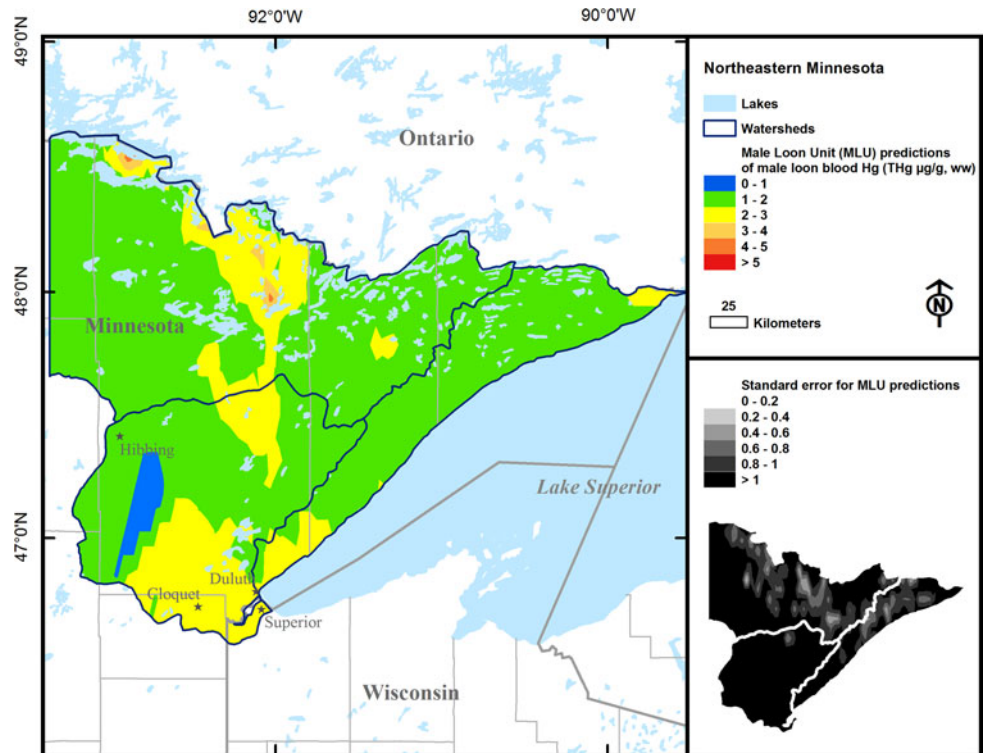
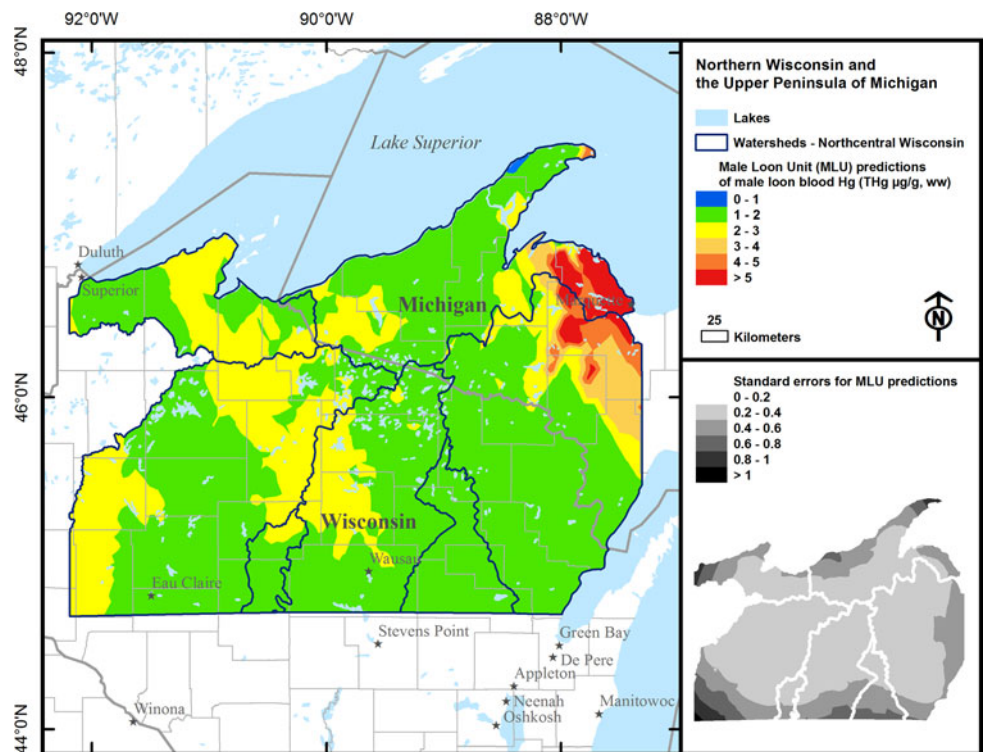


Fig. 4 Predicted male loon blood mercury values ($\mu\text{g/g}$ of total Hg in blood, ww) for a focal area in northern Wisconsin and the Upper Peninsula of Michigan, based on common loon and yellow perch Hg data. Inset map shows standard errors of the model-based predictions across the region



uncertainty associated with it. In contrast, in northern Wisconsin (Fig. 4) there is a central area where prediction standard errors are quite small, and thus the data may be interpreted with a higher degree of certainty. The north-eastern Minnesota region also illustrates the possibility for

additional analysis of the geospatial data; a review of the waterbodies containing the 798 Hg concentrations in loons and perch in the region indicate that over 25% of MLUs $>3.0 \mu\text{g/g}$, ww were found in reservoirs, including those with well documented elevated Hg concentrations.

Fig. 5 Predicted male loon blood Hg concentrations ($\mu\text{g/g}$ of total Hg in blood, ww) for a focal area in southern Ontario, based on common loon and yellow perch Hg data. Inset map shows standard errors of the model-based predictions across the region. Dotted areas are designated as agriculture ecumene (e.g., primarily agricultural zones) in Canada's 2006 Census of Agriculture (Statistics Canada 2006)

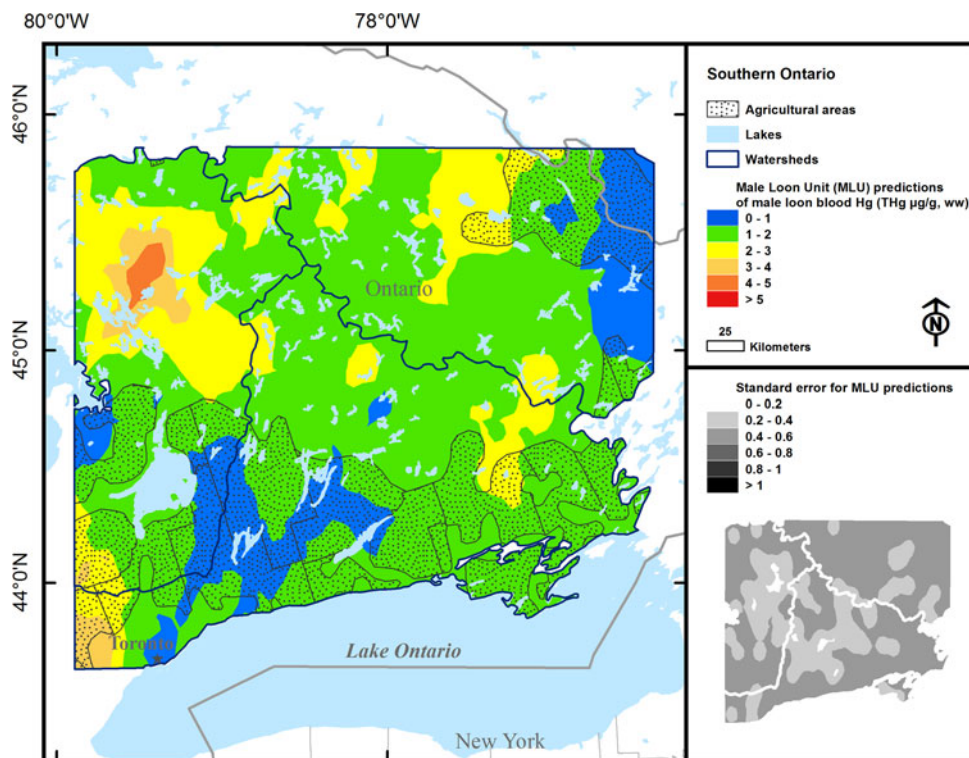
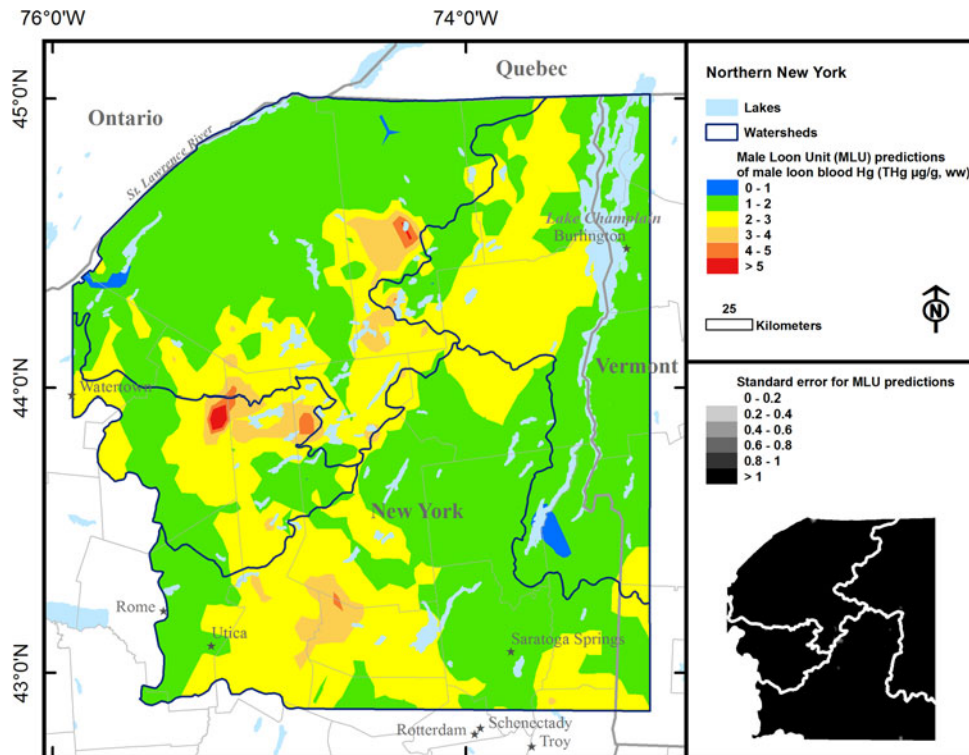


Fig. 6 Predicted male loon blood Hg concentrations ($\mu\text{g/g}$ of total Hg in blood, ww) for a focal area in northern New York, based on common loon and yellow perch Hg data. Inset map shows standard errors of the model-based predictions across the region; low predictive ability indicates highly heterogeneous focal area



The focal area with the greatest average MLU was northern New York ($2.38 \pm 1.43 \mu\text{g/g}$). Northern Wisconsin–Upper Peninsula of Michigan ($2.02 \pm 2.03 \mu\text{g/g}$)

and northeastern Minnesota ($1.92 \pm 1.01 \mu\text{g/g}$) both had average MLU values above the regional average of $1.80 \mu\text{g/g}$, and the lowest average MLU from the four areas

was southern Ontario ($1.46 \pm 0.86 \mu\text{g/g}$). The Wisconsin-Michigan and Minnesota site MLUs were not significantly different (Wilcoxon pairwise comparison, $\alpha = 0.05$, $P = 0.90$), but the Ontario and New York site MLUs were each significantly different from the other three sites ($P \leq 0.0001$). While roughly 70% of MLU values from southern Ontario were above $1.0 \mu\text{g/g}$, almost 94% of MLUs from northern New York were above. The highest FLUs and MLUs, both for the entire region and for the focal areas (Tables 1, 2), were based on yellow perch data, rather than loon data, and perch Hg data were highly variable. However, MLU averages for focal areas based on perch data versus loons were similar and did not vary in a consistent direction (e.g., average MLU estimates based solely on perch data were not consistently larger than MLU estimates based solely on loon data). Only compiled estimates, including both loon and perch data, are presented in Tables 1 and 2.

Discussion

The use of a standard metric that combines multiple tissues of a high profile species and its primary diet provides a robust dataset for understanding spatial gradients of MeHg availability for aquatic-dominated landscapes across much of the Great Lakes region. On a given water body, MLUs average higher Hg concentrations than FLUs because male loons average 21% larger than females (Evers et al. 2010). And, larger loons proportionally prey on larger fish (Barr 1996) and increasing fish size corresponds with higher Hg concentrations within a given species (Sandheinrich and Wiener 2011). Therefore, the largest loons on lakes with relatively elevated MeHg availability, where individual loons cannot fully depurate or demethylate dietary MeHg accumulated over multiple years (Evers et al. 1998), are likely the ones at greatest risk of adverse impacts to their behavior, physiology, reproduction and survival. However, a geographic examination of Hg profiles in the environment necessarily generalizes complex patterns in order to produce broadly applicable results. It is known, for example, that biological Hg contamination can vary widely even within a single water body, due to differences in landscape sensitivity, point sources, demethylation potential, and other factors. The type of large-scale spatial analyses conducted in this study subsume such landscape-level and habitat-related factors, but also occasionally obscure these relationships in favor of broader geographic patterns that are emphasized herein. As such, patterns of Hg accumulation in wildlife may need to be examined differently in this broad geographic framework, as in this case where loon body weight was not incorporated into regression models.

In addition, it is important to recognize that several major assumptions are necessary for the type of broad-scale spatial analysis conducted here, such as that of a common diet of loons across their range (e.g., including a similar proportion of similarly sized yellow perch, for example). Such assumptions, while reasonable based on the existing literature (Barr 1996; Evers et al. 2004; Burgess and Hobson 2006), have not been rigorously evaluated on a larger spatial scale. There are many factors which could influence the accuracy of this spatial analysis across the Great Lakes region, both locally and regionally, and in the absence of large-scale, standardized monitoring efforts, these spatial gradients of biological mercury do contain uncertainty. Nevertheless, the authors feel that this approach is useful for examining broad-scale trends and potential population-level impacts.

Biological mercury hotspots

Atmospheric deposition is often a primary pathway for anthropogenic Hg to enter an ecosystem (Gratz et al. 2010) and serves as a principal source for aquatic biota (Hammerschmidt and Fitzgerald 2006; Wiener et al. 2006). There is a growing body of evidence indicating that atmospheric deposition and biological Hg hotspots can be created by local emission sources, particularly when nearby areas are sensitive to Hg input. Three such studies include: (1) Steubenville, Ohio, where Keeler et al. (2006) demonstrated that nearly 70% of the Hg collected at a monitoring site originated from a neighboring coal-burning facility; (2) northeastern Massachusetts, where Hutcheson et al. (2008) reported a 32% decline in yellow perch Hg during a seven year decline in nearby Hg emissions from municipal and hospital incinerators; and (3) a study of southeastern New Hampshire lakes, where Hg concentrations declined over 50% in the blood of the common loon between 2001 and 2004 (Evers et al. 2007), and correlated with the removal of 6,600 lbs ($\sim 3,000 \text{ kg}$) of Hg from upwind incinerator emission sources located within 200 km of the study area. If these examples are not exceptions, then local emission sources can cause significant local impacts. With relatively high densities of Hg emission sources (e.g., coal-fired electric generators, incinerators, cement manufacturing facilities), the eastern US is expected to have relatively elevated concentrations of atmospheric deposition and therefore a greater potential for environmental Hg loads to create ecological effects. The recent identification and characterization of biological Hg hotspots in the northeastern United States and eastern Canada (Evers et al. 2007) demonstrated that proximity to local emission sources as well as landscape biogeochemical cycling were important factors. Similar explanations are valid for some areas within the Great Lakes region.

The degree of landscape sensitivity, often related to physicochemical and watershed attributes and atmospherically deposited Hg (or waterborne-oriented Hg), is critical information for understanding MeHg generation and bioavailability and the creation of biological Hg hotspots (Driscoll et al. 2007; Evers et al. 2007). Watershed features, including the percentage of wetlands, forest canopy and agriculture, drainage area, and slope can be used to predict piscivorous fish and bird Hg body burdens (Shanley et al. submitted.). Seven grids with MLUs averaging $>3.0 \mu\text{g/gHg}$, ww were identified in the Great Lakes Region (Fig. 2). Rationales for these biological Hg hotspots have been identified for four focal areas in northeastern Minnesota, northern Wisconsin-Michigan's Upper Peninsula, southern Ontario, and northern New York (Table 3). Focal areas primarily represent the northern half of the study area and reflect the distribution and density of relevant biological Hg concentrations. This analysis does not adequately characterize the southern Great Lakes region.

Northeastern Minnesota

This focal area is characterized by mixed deciduous and coniferous forests (79%) interspersed with scrub-shrub and emergent wetland, including open sphagnum bogs (12%), adjacent to natural and dammed waterbodies (8%) (Table 3). Reservoirs are an important feature of the landscape, while agriculture and urban areas are minimally present (1%). Although the underlying mechanisms responsible for enhancing Hg methylation, generation, and mobilization in reservoirs with water level fluctuations remain largely undefined, the influence of shoreline substrates experiencing regular wet-dry cycles is likely germane to the magnitude of methylation and subsequent bioavailability. The literature is replete with findings that fish, birds and other biota of newly created reservoirs are more contaminated with Hg than biota of comparable reference conditions (Bodaly et al. 1994; Anderson et al. 1995; Kelly et al. 1997; St Louis et al. 2004; Kamman et al. 2004), but it is the continuing water level fluctuations that are likely the most important process for enhancing MeHg availability to biota in most modern day reservoirs of the Great Lakes region. Over 25% of MLUs $>3.0 \mu\text{g/g}$, ww were found in reservoirs, including those with well documented elevated Hg concentrations (Crane, Little Vermillion, and Sand Point Lakes; Sorensen et al. 2005). Another grouping of waterbodies with elevated MLUs includes relatively small natural lakes with low water pH and elevated dissolved sulfate and total organic carbon. Some of these lakes are located in Voyageurs National Park (Wiener et al. 2006) and others are found within the Superior National Forest.

Table 3 Factors contributing to mercury concentrations at four focal areas in the Laurentian Great Lakes region

Focal area	State/ province	Wetlands (%)	Water (%)	Forest (%)	Agriculture/ forbland (%)	Urban (%)	Hypothesized mechanisms responsible for biotic Hg concentrations				
							Water-level management	Landscape sensitivity (elevation and/or pH)	Local air emissions	Local soil/water contamination	Biodilution
Northeastern Minnesota	Minnesota	11.5	8.3	78.8	0.8	0.6	X	-	-	-	-
Northern Wisconsin and the Upper Peninsula	Wisconsin, Michigan	19.5	2.7	61.1	15.8	0.9	-	X	-	-	X
Southern Ontario	Ontario	0.3	5.9	73.1	18.8	1.9	-	X	X	-	X
Northern New York	New York	5.9	5.4	73.4	13.0	2.3	-	X	X	-	-

Degree of landscape sensitivity, often related to physicochemical and watershed attributes and atmospherically deposited Hg (or waterborne-oriented Hg), is critical information for understanding MeHg generation and bioavailability and the creation of biological Hg hotspots (Driscoll et al. 2007; Evers et al. 2007). Percent land cover in wetlands, forest canopy, and agriculture are landscape-level characteristics that have been found to influence Hg exposure in wildlife (Shanley et al., submitted)

Northern Wisconsin–Upper Peninsula of Michigan

This focal area is characterized by mixed deciduous and coniferous forests (61%) interspersed with scrub-shrub and emergent wetland (20%) that are often adjacent to natural and dammed waterbodies (3%) (Table 3). In this focal area, reservoirs are not prevalent, while agriculture and urban areas are interspersed at a level that is relatively higher than the other focal areas. Intensive research within this focal area, particularly in northern Wisconsin, has resulted in a highly defined environmental Hg exposure gradient, both spatially (demonstrated in the low standard errors of the model-based spatial predictions (Fig. 4), and temporally (Meyer et al. 2011). Field measurements of lake chemistry and fish and loon Hg concentrations for this area indicate that elevated Hg concentrations are primarily driven by low alkalinity (Cope et al. 1990; Meyer et al. 1995, 1998). The complexity of physicochemical processes, which include factors such as water pH, sulfate, and dissolved organic carbon, are also responsible for in-lake Hg methylation (Watras and Morrison 2008). Because of the intensive and standardized investigations recording loon Hg concentrations over a relatively extended period of time, this focal area provides one of the better locations to examine temporal trends of biotic Hg concentrations within the Great Lakes region. Meyer et al. (2011) found through a linear mixed effects model that loon blood Hg concentrations declined from 1992 to 2000 and increased during 2002–2010—a biphasic response. While a similar biphasic response was found for piscivorous fish in Minnesota (Monson 2009) and to a lesser extent for piscivorous fish in Lake Erie (Bhavsar et al. 2010), environmental Hg loads in Wisconsin piscivorous fish were spatially stratified. Southern parts of the state annually increased 0.8%, while northern Wisconsin exhibited an annual decline of 0.5% (Rasmussen et al. 2007).

A review of the water bodies in this focal area, containing 1,687 Hg concentrations in loons and perch, indicate that all predicted MLUs >3.0 $\mu\text{g/g}$, ww were located in northwestern Wisconsin and a small northern area in the western Upper Peninsula of Michigan (Fig. 4). While only small numbers of scattered reservoirs or flowages are present, these waterbodies often are the ones with elevated MLUs. The grid meeting the criteria of a biological Hg hotspot (Fig. 2) includes waterbodies with elevated MLUs that have both natural and dammed waterbodies in close proximity to contaminated soil and water point sources. It is well described, that some waterbodies like Deer Lake, Michigan have highly contaminated aquatic biota because of legacy Hg-laden waste disposal that was co-located with copper mining and smelting sources (Kerfoot et al. 1999).

Southern Ontario

This focal area is characterized by mixed deciduous and coniferous forests (73%) interspersed by inconsequential areas of wetlands ($<1\%$) and natural and dammed waterbodies (6%) (Table 3). In this focal area, agriculture and urban areas (20%) are a relatively important component of the landscape and are primarily restricted to the southern half. The biological Hg hotspot identified from the dataset includes an area investigated by Rasmussen et al. (1998) using lake sediment cores; in this case on-site observations indicated that sediment and fish Hg concentrations could be related to natural sources, although the ability of aquatic ecosystems to use atmospherically deposited Hg in methylmercury production and availability cannot be eliminated. Also, numerous lakes in the area studied by Rasmussen (1998) and eastward are low alkalinity, low pH water bodies, conditions well-known to be associated with elevated levels of Hg in fish and common loons in south-central Ontario (Scheuhammer et al. 1998). Based on an assessment of Hg concentrations in small fish from a provincial government database, Scheuhammer and Blancher (1994) suggested that up to 30% of Ontario lakes have small fish (<250 g) with Hg concentrations exceeding thresholds for reproductive impairment in common loons. The Canadian Shield has a greater capacity for piscivorous fish and wildlife to contain elevated Hg body burdens than areas south of it, such as much of this study's focal area.

While the potential underlying mechanism for the biological Hg hotspot is challenging to determine, this focal area does provide an opportunity to relate lower than average MLUs (Table 2) to environmental factors such as a high proportion of alkaline high-pH lakes in more southern areas of the province, as well as the elevated use and release of phosphorus and nitrogen related to agriculture and urban uses. When a large degree of primary and secondary production is evident, the ability of MeHg to biomagnify through the upper trophic levels is inhibited—or bloom dilution occurs (Pickhardt et al. 2002; Chen and Folt 2005). This is due to a reduction in biomagnification of MeHg to top-level consumers when the pool of MeHg is biodiluted across a larger amount of biomass in the planktonic trophic concentrations. This concept was experimentally demonstrated by measuring Hg bioaccumulation across a nutrient enrichment gradient in a large mesocosm experiment (Pickhardt et al. 2002), and subsequently validated by field surveys in more than 150 lakes in the Northeast US (Kamman et al. 2004; Chen and Folt 2005). In the mesocosm experiment, nutrient additions and resulting algal blooms decreased the mass-specific MeHg concentrations in algae and in zooplankton. Mercury concentrations in predatory fish were strongly influenced by

the size and structure of zooplankton (Stemberger and Chen 1998; Chen et al. 2000).

Northern New York

This focal area is characterized by mixed deciduous and coniferous forests (73%) interspersed with scrub-shrub wetland, including open sphagnum bogs (6%), adjacent to natural and dammed waterbodies (6%) (Table 3). In this focal area, agriculture and urban areas (15%) are interspersed and primarily are located around the periphery of this focal area. Lakes in this region are characterized by low dissolved organic carbon (<5 mg C/l), low pH (<6.3), and low acid neutralizing capacity (ANC <100 ueq/l) (Yu et al. 2011); these lake physicochemical properties appear to increase bioavailability of Hg at the base of the aquatic food web (Adams et al. 2009; Dittman and Driscoll 2009; Dittman et al. 2009). Watershed features, such as percent wetland adjacent to waterbodies, also appear to be important drivers for MeHg transfer through aquatic food webs into fish (St Louis et al. 1994) and avian piscivores, such as the common loon (Kramar et al. 2005). Because this region, particularly the area within Adirondack State Park, is sensitive to Hg inputs it has the highest average MLU Hg concentration of the four focal areas; over 20% of the loon population is estimated to be at potential risk to significant losses in reproductive abilities (Table 2). Two biological Hg hotspots are present in northern New York and both are related to groups of small lakes with physicochemical features identified as generating high MeHg availability. Some of these lakes are at relatively high elevations—a landscape feature that also contributes to elevated Hg deposition, which may be linked to increased MeHg availability (Rimmer et al. 2005). However, because of the high standard error for this focal area, likely reflecting tremendous heterogeneity in spatial Hg patterns, predictive abilities are limited using general spatial models. In response, lake-specific assessments based on water, fish and loon Hg concentrations are being developed for evaluating adverse ecosystem effects (Simonin et al. 2008; Yu et al. 2011).

Projected regional ecosystem impacts

The ability to link atmospheric deposition to aquatic biota, identify and characterize the presence of biological Hg hotspots, and model injury from environmental Hg loads to avian piscivores at a regional level are important steps for better understanding ecological problems related to MeHg toxicity. While most air, soil and water point sources are distributed across the southern Great Lakes, this study was limited to examining the spatial gradient of environmental Hg in the northern Great Lakes (Fig. 2). Spatial analyses indicate a broad range of environmental Hg concentrations

that are driven by not only local atmospheric deposition and soil or water contaminations (e.g., Onondaga Lake, New York; Wang and Driscoll 1995), but hydrological, physicochemical, and watershed features as well. The wide distribution of biological Hg hotspots likely reduces the potential adverse impacts at a regional level for avian piscivores. However, some areas have greater risk than others, and they include three of the focal areas: northeastern Minnesota, north-central Wisconsin–Upper Peninsula of Michigan, and northern New York (Fig. 2).

Overall, 3.7% of the FLUs and 9.8% of the MLUs exceed the established LOAEL. In some focal areas, MLUs exceed the LOAEL in over 20% of the breeding population. When such a high proportion of the breeding population is above the LOAEL, mercury exposure may have the potential to cause population-level impacts (Evers et al. 2004; Evers et al. 2008; Burgess and Meyer 2008). However, this is a conservative estimate of adverse impact, as the reproductive harm of Hg body burdens above the NOAEL of 1.0 µg/g, ww is not included. Over 72% of the MLUs across the northern Great Lakes Region are above the NOAEL and below the LOAEL. Further definition describing the magnitude of adverse impacts from MeHg toxicity on loons and other avian piscivores remain as a high priority need for the Great Lakes region.

Policy and management implications

In this study, the majority of elevated MLU values are from state and federal public lands (Fig. 1). Given the potential for population-level effects from MeHg exposure, determination of the specific sources of Hg affecting upper trophic level aquatic biota in the Great Lakes is needed and could greatly inform governmental policy and regulatory decisions. Widespread, and often elevated, biological Hg exposure implies the need for Hg monitoring at a regional and national scale that can track changes in Hg emissions and effluents. Long-term Hg monitoring across the Great Lakes region by the Mercury Deposition Network needs to be supplemented with components as described in the proposed National Mercury Monitoring Program or MercNet (Mason et al. 2005; Harris et al. 2007; USEPA 2008; Schmeltz et al. 2011). Such a standardized network is necessary to inform federal and state Hg-related policies, provide data for predictive models, and characterize the biological effects in the United States from the redistribution of anthropogenic Hg on the landscape. In Canada, the CARA program is actively contributing to this need (Morrison 2011). In addition to the development of a large-scale United States monitoring network, there are recent advances in the study of Hg in the environment that could further contribute to our understanding of Hg contamination in the Great Lakes region. Small variations in the stable,

naturally occurring isotopic composition of Hg can be used to trace sources of environmental Hg contamination and assess the role of locally produced Hg in affecting sensitive ecosystems within the Great Lakes region (Bergquist and Blum 2007; Blum and Bergquist 2007). The availability of such new techniques means that natural resource managers may, for the first time, have the predictive capability to proportionally link specific Hg point sources with remote contamination in fish and wildlife across neighboring landscapes.

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