

Spatiotemporal trends of mercury in walleye and largemouth bass from the Laurentian Great Lakes Region

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Abstract The risk of mercury (Hg) exposure to humans and wildlife from fish consumption has driven extensive mercury analysis throughout the Great Lakes Region since the 1970s. This study compiled fish-Hg data from multiple sources in the region and assessed spatiotemporal trends of Hg concentrations in two representative top predator fish species. Walleye (*Sander vitreus*) and largemouth bass (*Micropterus salmoides*) were chosen for the trend analysis because they had more Hg records (63,872) than other fish

species that had been sampled from waters throughout the region. Waterbody types were inland lakes (70%), the Great Lakes, impoundments, and rivers. The compiled datasets were analyzed with a mixed effects statistical model having random effects of station, year, and fish length; and fixed effects of year, tissue type, fish length, habitat, and season. The results showed a generally declining temporal trend in fish-Hg for the region (1970–2009), with spatial trends of increasing Hg concentration from south to north and from west to east across the region. Nonlinearity was evident in the general downward trends of Ontario walleye, with a shift to an upward trend beginning in the 1990s. Only ongoing monitoring can reveal if this upward shift is an oscillation in a long-term decline, a statistical anomaly, or a sustained declining temporal trend in regional fish-Hg concentrations.

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Introduction

Mercury (Hg) has been monitored in Great Lakes Region (GLR) fish as a part of multiple state, provincial, tribal, and federal contaminant monitoring programs. Many programs began in the 1970s or early 1980s. As anthropogenic releases of Hg change over time—decreasing regionally (e.g., Europe and North America) but increasing globally (Pacyna et al. 2006)—there is a need to understand how this affects trends in fish-Hg concentrations. When Hg deposition declines, we expect to see declines in fish-Hg concentrations; however, the anticipated response time, and its possible variation among regions, are unclear. Complicating the response are probable changes in the

watershed transport, biogeochemistry, and bioavailability of Hg over time resulting from ecosystem alterations in the GLR (Munthe et al. 2007a). In particular, documented increasing temperature in the Great Lakes (Austin and Colman 2008), and other factors related to changing climate are sufficient forcing factors to increase Hg bioavailability despite reductions in Hg deposition as suggested by recent Canadian arctic studies (Carrie et al. 2010; Kirk et al. 2011).

Previous studies have examined trends in fish-Hg concentrations within subsets of the compiled GLR data in this study. Trend analysis of walleye (*Sander vitreus*) in Wisconsin lakes between 1982 and 2005 has shown an annual percent change (APC) of 0.5 to 0.6% in northern Wisconsin and an upward trend of 0.8% APC in southern Wisconsin (Madsen and Stern 2007; Rasmussen et al. 2007). Over the same period in Minnesota (1982–2006), the overall trend in walleye and northern pike (*Esox lucius*) was the same as northern Wisconsin, but a better fitting piecewise linear regression model indicated a steeper decline of 4.7% per year from 1982 to 1992, followed by an increase of 1.4% per year for 1992–2006 (Monson 2009). A similar downward overall trend of 0.5–1% per year has been observed in five fish species from the Hudson River, New York between 1970 and 2004 (Levinton and Pochron 2008) and in yellow perch (*Perca flavescens*) from New York lakes (Simonin et al. 2009). In the Great Lakes, Hg in lake trout (*Salvelinus namayush*) has generally declined over the last 30 years; however, recent trends in walleye have been flat in Lake Ontario and increasing in Lake Erie (Bhavsar et al. 2010). The leveling off of the trend in Hg concentrations was observed for multiple species from Lake St. Clair (Gewurtz et al. 2010). A U.S. national dataset of multiple fish species found some upward trends but overall the Hg trend was downward from 1969 to 2005 (Chalmers et al. 2010).

The objectives of this study were (1) to compile fish-Hg data from multiple sources in the GLR into a database for ongoing descriptive statistics and hypothesis testing, (2) to assess spatiotemporal trends of Hg concentrations in two fish species that best represent the GLR, and (3) to compare trends among sub-regions of the GLR.

Materials and methods

Compiled data

Data were compiled primarily from state, tribal, and provincial government contaminant monitoring programs, which included the eight Great Lakes states and the Province of Ontario (Ontario Ministry of the Environment (OMOE). Environment Canada (EC) provided a dataset

from their Fish Contaminants Monitoring and Surveillance Program (FMSP) and the U.S. Environmental Protection Agency's Great Lakes National Program Office (GLNPO) provided data from their Great Lakes FMSP. The Great Lakes Indian Fish and Wildlife Commission (GLIFWC) provided a dataset from lakes in the ceded territories located in Wisconsin, Minnesota, and Michigan. BioDiversity Research Institute compiled all datasets, except OMOE, into a single dataset, "MercNet," with a common sample and site identification. OMOE data were analyzed separately from MercNet because of a different database structure. To compare results for the two datasets, an "M" for MercNet and "O" for OMOE are affixed to the fish species abbreviations. All Hg concentrations are reported as total Hg on a wet-weight basis.

The Great Lakes states monitoring program follows the Great Lakes protocol for consumption advisories, which stipulates skin-on fillets (Anderson et al. 1993). As with the state monitoring programs, GLIFWC and OMOE collect fish contaminant data primarily to provide fish consumption advisories. Unlike the other programs, GLIFWC and OMOE use skin-off fillets as their standard tissue sample. Other tissue types (e.g., whole fish) are considered by OMOE for special studies. Both of the EC and USEPA datasets are primarily whole fish homogenates. A trend-monitoring program in Michigan also relies on whole fish samples. The period of record for each dataset ranged from

Table 1 Database providers, period-of-records, and number of Hg records for largemouth bass and walleye in the Great Lakes region

Data provider	Period of record	Hg records
Great Lakes Indian Fish and Wildlife Commission	1989–2008	4,803
Environment Canada	1977–2008	1,221
US Environmental Protection Agency	2004–2008	46
Illinois Environmental Protection Agency	1985–2008	628
Indiana Department of Environmental Management	1983–2006	426
Michigan Department of Environmental Quality	1984–2007	4,349
Minnesota Pollution Control Agency	1967–2007	7,137
New York Department of Environmental Conservation	1970–2008	3,558
Ohio Environmental Protection Agency	1970–2007	911
Pennsylvania Department of Environmental Protection	1979–2009	216
Wisconsin Department of Natural Resource	1977–2007	7,501
MercNet Hg records for both species		30,796
Ontario Ministry of the Environment (analyzed separately from MercNet data)	1970–2009	33,326

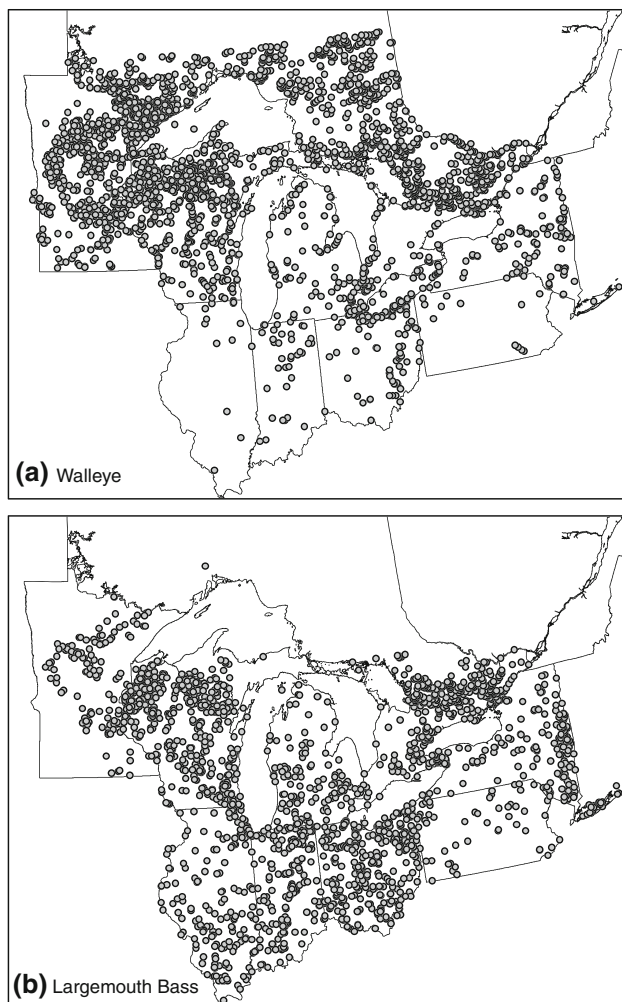


Fig. 1 Spatial distributions of combined sites from MercNet and OMOE datasets for **a** walleye and **b** largemouth bass

the 1970s or early 1980s and continued to 2005–2009 (Table 1).

The compiled MercNet database for the GLR contains 105,287 records of Hg concentrations with samples for 240 species and 37 tissue types. The OMOE database has more than 200,000 Hg records; with samples for 100 species and 15 tissue types. Both datasets include some non-fish species, representing about one percent of the samples. For this study, walleye and largemouth bass (*Micropterus salmoides*) were chosen for the trend analysis because they were the fish species with the greatest number of samples in the datasets and their sample locations were distributed throughout the GLR (Fig. 1). Walleye made up 72% of the 30,796 records for these two species from MercNet and 86% of the 33,326 from the OMOE dataset (Table 2).

Northern pike (*E. lucius*) and lake trout (*Salvelinus namaycush*) are common indicator species for fish-Hg concentrations but were not considered for this study. Northern pike is the most common species in the

Minnesota database and was the primary species in a previous fish-Hg trend analysis (Monson 2009). The MercNet database contained 66% more northern pike than largemouth bass; however, 98% of the northern pike were in Michigan, Minnesota, and Wisconsin. By comparison, 52% of the largemouth bass were in those three states, providing a broader spatial coverage. Lake trout have also been a typical common species for contaminant trends in the Great Lakes, but were not used as an indicator in our study because they represented a relatively small number of monitoring stations for the GLR as a whole.

The analysis conducted in this study included categorical variables for waterbody type and tissue type. The MercNet database included a field for waterbody type and each site was categorized as a lake, impoundment, river, Great Lake, or “other.” The last category included canals, channels, lagoons, harbors, and marsh. The OMOE data were classified in lake/impoundment, river, or Great Lake categories. Tissue type included three groups: skin-on fillet, skin-off fillet, and whole fish. Skin-on fillets comprised 73% of the tissue types in MercNet (Table 2). Most of the state fish contaminant monitoring programs followed the Great Lakes protocol for sport fish consumption advisories, which stipulates skin-on fillets as the standard sample tissue type (Anderson et al. 1993); MercNet walleye samples were 65% skin-on and 25% skin-off fillets. MercNet largemouth bass were 93% skin-on and only 1% skin-off fillets. Whole fish samples, which have been used mostly for assessing temporal trends, ecosystem status, and risks to fish and fish-consuming wildlife, accounted for only 8.6% of the total records for walleye and largemouth bass in MercNet. For this study, 65% of the whole fish were walleye from the Great Lakes and 54% of the whole fish records were from Lake Erie. Environment Canada’s monitoring program accounted for 1,221 (48%) of the whole fish records and Michigan’s trend monitoring sites accounted for 952 (38%). The remaining 14% were from multiple state monitoring programs. The dataset from OMOE had skin-off fillets as the standard portion, which represented 99.7% of the walleye and largemouth samples in the dataset.

Because this was a regional analysis of trends, each site was considered representative of its particular subregions; therefore, sites contaminated by local point sources did not fit this regional representation criterion. In addition to industrial point sources, such as chlor-alkali plant discharge, there was a concern that some river and impoundment sites received treated wastewater from facilities that used Hg-based slimicides prior to controls being implemented beginning in the 1970s. The only contaminated site identified in the MercNet database was Onondaga Lake in New York. Onondaga Lake has a long monitoring record, from 1973 to 2008, with a total of 689 records. Hg concentrations in Onondaga walleye from the

Table 2 Sample counts by water-body type, species, and tissue-type in the databases provided by MercNet and OMOE

Table 2 Sample counts by water-body type, species, and tissue-type in the databases provided by MercNet and OMOE	Water-body type	Total	Species		Tissue type			
			WAE	LMB	FILET	FILSK	WHORG	WNOHV
	MERCNET							
	Lake	21,665	15,611	6,054	5,370	15,767	518	10
	Impoundment	3,542	2,410	1,132	188	3,277	71	6
	River	3,094	1,844	1,250	39	2,640	294	121
	Great Lake	2,352	2,308	44	18	701	1,633	0
	Other	143	19	124	0	137	2	4
	Total	30,796	22,192	8,604	5,615	22,522	2,518	141
OMOE								
	Lake & impoundment	22,759	19,161	3,598	22,691	50	18	0
	River	5,170	4,531	639	5,152	15	3	0
	Great Lake	5,397	4,897	500	5,397	0	0	0
	Total	33,326	28,589	4,737	33,240	65	21	0
	MERCNET-OMOE total	64,122	50,781	13,341	38,855	22,587	2,539	141

WAE walleye, LMB largemouth bass, FILET skin-off fillet, FILSK skin-on fillet, WHORG whole fish, WNOHV whole fish except head & viscera
Other includes canals, channels, harbors, lagoons, and marsh
WNOHV were not used in the statistical model

1970s were as high as 7.91 mg/kg. In the mid-1990s, monitoring switched primarily to largemouth bass. Hg concentrations declined after the 1970s, but continued to exceed 3.0 mg/kg. All Onondaga records were excluded from the database for development and testing of the statistical model because of the lake's well-documented Hg pollution. Other sites are known to have point source contamination of Hg, such as Deer lake in Michigan, Sandpoint lake in Minnesota, Dunham Reservoir and Franklin Falls Flowage in New York, and Wisconsin River and Fox River in Wisconsin. Outlier sites were identified through exploratory data analysis, such as distribution plots, and removed before applying the statistical model. Individual records of high Hg concentration were not removed. Only Onondaga was identified as an outlier site using these methods. The record counts from the other contaminated sites were relatively small and had a wide range of Hg concentrations; therefore, they were not removed from the dataset.

Statistical analysis

The objective of the descriptive statistical model was to estimate the temporal and spatial trends in Hg concentrations while accounting for multiple variables that influence fish-Hg levels in sample tissues. The primary statistical tool was a mixed effects linear model (Kutner et al. 2005), which contained both fixed and random effects:

$$\log_e(\text{Hg}) = (\beta_0 + b_{0Y} + b_{0S}) + (\beta_1 + b_{1S}) * Y + (\beta_2 + b_{2Si}) * L + T + \beta_3 * L * T + W + D + \varepsilon$$

where $\beta_0, \dots, \beta_3$ were fixed effect regression parameters, Y = sample year, L = fish total length, T = sample tissue type, W = waterbody type, D = sample season, and ε is

residual error distributed as Normal(0, σ); the fixed effect trend, β_1 , represents the average linear trend in Hg levels across all GLR sites during the study and is a primary parameter of interest in the analysis. The random effects were used to account for correlations among tissue samples taken within the same year or at the same sampling site (inland lakes are single sites; each of the Great Lakes have multiple sites); b_{0Y} represented sample year effects (i.e., correlation in Hg levels among all samples taken in a given year) distributed as $N(0, \sigma_Y)$; b_{0S} represented site effects (i.e., correlations in Hg for samples from the same site) distributed as $N(0, \sigma_{0S})$; b_{1S} represented site deviations from the fixed trend distributed as $N(0, \sigma_{1S})$, which allows each site to have a unique temporal trend; and b_{2S} were site-specific deviations from the overall Length-Hg relationship distributed as $N(0, \sigma_{2S})$. A slightly different mixed model was applied to OMOE dataset, using a single tissue type, skin-off fillet, which represented nearly 100% of the samples.

The period of record for this analysis was 1970–2009 (39 years). To enhance interpretability, the fish length data were centered at the mean fish length, and the year variable was centered at 1995 for MercNet and 1990 for OMOE. Centering the year variable at its average value decreases correlation between estimates of intercept and slope. The β_0 parameter is the overall intercept, and describes the average Hg concentration for a fillet from an average length fish caught in a GLR lake in spring of the centered year. Other fixed parameters were incorporated into the model to control for factors likely to affect fish-Hg sample concentrations such as fish length, sample type, waterbody (i.e., habitat) type, and season of capture.

Though the random year effect, b_{0Y} , and station effects on Hg trend, b_{0S} and b_{1S} , are strictly variance components in the analysis, the model provided unique predictors for the realized random effects. These predictors are denoted as BLUPs

for ‘best linear unbiased predictors’, and were used to determine annual deviation from the linear trend, to estimate Hg levels and trends for individual lakes (or Great Lakes sampling stations), and to examine regional differences. For example, the terms $(\beta_0 + b_{0Y1995} + b_{0Si})$ give the expected $\log_e$ fish-Hg concentrations for a tissue sample taken from an average length fish in the i th site in spring of the centered year, and the terms $(\beta_1 + b_{1Si})$ give the predicted trend in $\log_e$ fish-Hg concentrations for site i . The original GLR fish-Hg model for MercNet walleye included spatial parameters (latitude and longitude), but they were removed from the model after deciding there was a likely temporal sampling bias caused by more recent sampling of lakes with high Hg concentrations in the eastern GLR (i.e., New York). Instead of including the spatial parameters in the model, a revised approach looks at the estimated station effect through BLUPs.

Fish age and gender are also variables that can affect fish-Hg, although they were not included in the GLR fish-Hg model because they were not available for many records. Age is generally represented by fish length because determining age accurately is time consuming and difficult in older individuals. Gender is usually not recorded on most samples; it may not be grossly apparent during all seasons and for all sizes of fish.

Descriptive statistics and graphics were prepared with SYSTAT (V. 13) and SigmaPlot (V. 11) (SYSTAT Software, Inc. 2009), as well as ArcGIS (ESRI ArcMap™ V. 9.3.1, 2009). Statistical modeling was performed in R (R Development Core Team 2010) using the lme4 package.

Results and discussion

Descriptive Statistics

The distribution of fish lengths were symmetric on a linear scale (median and mean were nearly equal), indicating

length measurements were normally distributed (Fig. 2a). The distributions of Hg concentrations were skewed on a linear scale (medians less than means) because the concentrations were lognormally distributed (Fig. 2b). Therefore, for Hg concentrations, medians were the preferred measure of central tendency and the data were log-transformed for statistical analysis. Within both datasets, median fish lengths of largemouth bass were 10–11 cm less than those for walleye (Table 3). Median Hg concentrations for MercNet walleye and largemouth bass were not significantly different (0.32 and 0.31 mg/kg), whereas Ontario walleye and largemouth bass were significantly different at 0.38 mg/kg and 0.28 mg/kg, respectively.

Compared to fish-Hg concentrations in northeastern North America and Scandinavia—two comparable regions where large datasets of fish-Hg concentrations have been compiled—the records compiled for our study indicate that fish from the GLR accumulate Hg to lower average concentrations. Mean skin-on fillet concentrations in MercNet walleye and largemouth bass were 0.45 and 0.40 mg/kg, respectively (Table 3). In northeastern North America, mean Hg concentrations in fillets were 0.76 mg/kg in walleye (mean length 44.6 cm) and 0.54 mg/kg in largemouth bass (mean length 35.8 cm) (Kamman et al. 2005). In Scandinavia, the reported mean Hg concentration in standardized 1 kg northern pike filets was 0.73 mg/kg (Munthe et al. 2007b). The reason for disparity is unclear given the likely differences in multiple causal factors, such as Hg deposition, type of waterbodies, season of collection, or fish growth rate.

Mixed effects model

Output summary

Inputs for each species-dataset model presented previously are summarized in Table 4. The first set of output

Fig. 2 Statistical distributions of **a** fish total length and **b** Hg concentration for walleye (WAE) and largemouth bass (LMB) in the MercNet (M) and OMOE (O) datasets. Box plots show interquartile range (shaded area), median and mean (vertical solid and dashed lines within shaded area), 10th and 90th percentiles (end of whiskers), and 5th and 95th percentiles (open circles)

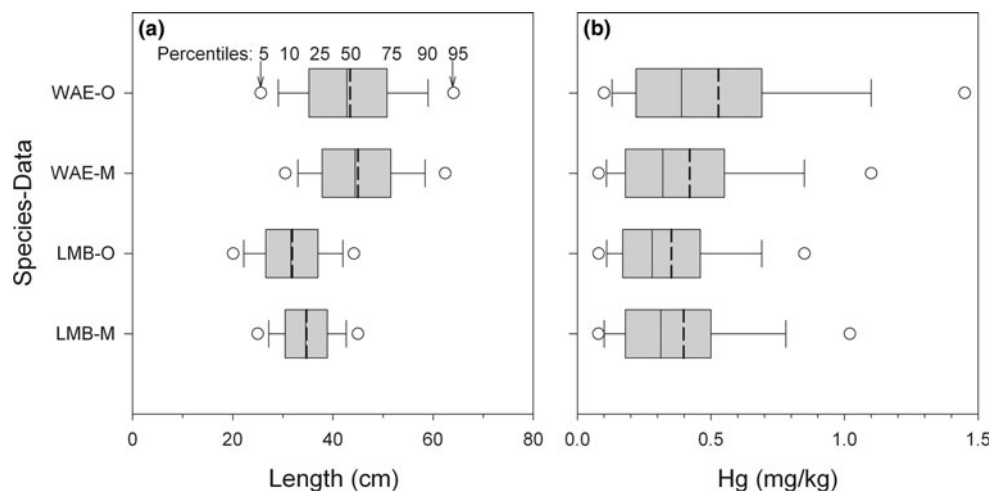


Table 3 Summary statistics of fish length and Hg concentrations for largemouth bass and walleye by tissue type

Species	Tissue	Total length (cm)					Hg (mg/kg)				
		<i>N</i>	Median	Mean	S.D.	S.E.	<i>N</i>	Median	Mean	S.D.	S.E.
<i>MERCNET database</i>											
LMB	FILSK	7,854	34.5	34.8	6.3	0.07	7,970	0.320	0.404	0.333	0.004
	FILET	83	34.3	34.7	6.1	0.67	104	0.319	0.351	0.218	0.021
	WHORG	383	36.0	33.9	7.1	0.36	399	0.230	0.284	0.207	0.010
	WNOHV	130	35.1	35.4	7.7	0.67	131	0.360	0.403	0.331	0.029
	All types	8,450	34.5	34.7	6.4	0.07	8,604	0.313	0.398	0.331	0.004
WAE	FILSK	14,370	44.4	44.8	10.0	0.08	14,552	0.340	0.449	0.398	0.003
	FILET	5,493	42.9	44.5	9.8	0.13	5,511	0.352	0.435	0.303	0.004
	WHORG	1,972	49.0	47.8	10.0	0.23	2,119	0.140	0.185	0.146	0.003
	WNOHV	10	36.5	37.8	6.3	2.01	10	0.260	0.379	0.278	0.088
	All types	21,845	44.5	45.0	10.0	0.07	22,192	0.320	0.420	0.367	0.003
<i>OMOE database</i>											
LMB	FILET	4,737	31.6	31.9	7.3	0.11	4,737	0.280	0.352	0.268	0.004
WAE	FILET	28,580	42.8	43.4	11.6	0.07	28,589	0.380	0.527	0.638	0.004

WAE walleye, LMB largemouth bass, *FILSK* skin-on fillet, *FILET* skin-off fillet, *WHORG* whole fish, *WNOHV* whole fish except head & viscera (WHNOV not used in statistical modeling)

information is a summary of variance components. Total and residual variance components were similar among the species datasets. More than 80% of the variance was explained for all four species datasets by the parameters considered.

A second set of outputs is the parameter estimates for continuous variables. The intercept is the mean Hg concentration of a mean length fish caught in the spring from a lake for the mean year of the dataset. For MercNet data, the intercept is for a skin-on fillet and the mean length is from the combined walleye and largemouth bass dataset. For OMOE data, the intercept was for skin-off fillets, because no skin-on fillets were included in the OMOE data. The specificity of each intercept precluded their direct comparison. BLUPs for intercepts of the same year, fish length, and tissue type (i.e., MercNet data adjusted for skin-off fillets) provide a better comparison. The intercept describes overall averages of Hg among lakes, whereas the BLUPs give lake-specific deviations from the overall average. For a skin-off fillet from a 42 cm fish in 2008, the BLUPs were 0.294 mg/kg for MercNet walleye, 0.405 mg/kg for OMOE walleye, 0.410 mg/kg for MercNet largemouth bass, and 0.483 mg/kg for OMOE largemouth bass. Higher Hg concentrations of the respective species in the OMOE dataset are further explored in the spatial trends section.

Largemouth bass at the same length as walleye are expected to be older and therefore have potentially higher Hg concentrations. A mean length of 42 cm corresponds to about the 50th percentile of 6-year old largemouth bass and a 4-year old walleye (Quist et al. 2003; Jackson et al. 2008). This and possibly other bioenergetic factors result in

generally higher Hg concentrations in largemouth bass compared to the same sized walleye (Bhavsar et al. this issue). Despite this difference at a given length, walleye live longer, reach larger sizes, and typically accumulate higher maximum Hg concentrations than largemouth bass (Schneider et al. 1977; Green and Heidinger 1994; Bhavsar et al. this issue). A comparison of same-age fish was not possible from these monitoring datasets because of the paucity of age data associated with fish length.

The positive linear relationship between log Hg concentration and fish length, which varied among species and waterbodies, is well documented. The effect of length in the mixed model was 4.6% per cm of fish length for both sets of walleye. Largemouth bass had significantly steeper length effects at 5.4% per cm in MercNet and 6.2% in the OMOE data. This difference in length effect between species is consistent with the observations that Hg concentrations are higher for largemouth bass compared to walleye at the same length (Bhavsar et al. this issue).

Year effect slopes corresponded for all four species sets, with mean annual percent declines in Hg of 0.6–0.9%. This downward trend agrees with previous statistical models applied to geographic portions of this dataset (Madsen and Stern 2007; Rasmussen et al. 2007; Monson 2009). The year effect slope is evaluated in more detail below under temporal trends.

Compared to inland lakes, Great Lakes Hg concentrations for walleye were 58 and 52% lower for MercNet and OMOE, respectively. The walleye from the Great Lakes were almost entirely from Lake Erie, which has the lowest Hg in fish among the Great Lakes (Bhavsar et al. 2010).

Table 4 Summary of inputs and outputs for the mixed effects model

Parameter	WAE-M	WAE-O	LMB-M	LMB-O
Input				
Year range	1970–2009	1970–2009	1970–2009	1970–2009
Years (<i>n</i>)	39	39	34	37
Stations (<i>n</i>)	2,202	927	1,959	316
Records (<i>n</i>)	21,358	28,487	7,728	4,701
Center year	1995	1990	1995	1990
Center total length (cm)	42.1	43.4	42.1	31.9
Output: residual and explained variance				
Total variance	0.640	0.700	0.621	0.542
Residual variance	0.106	0.114	0.078	0.104
Percent variance explained	83%	84%	87%	81%
Output: continuous variables (linear units and 2 S.E. range)				
Intercept (mg/kg)	0.310 (0.296, 0.324)	0.487 (0.457, 0.519)	0.427 (0.404, 0.451)	0.308 (0.283, 0.335)
Total length (Pct/cm)	4.55 (4.45, 4.66)	4.58 (4.48, 4.68)	5.38 (5.16, 5.61)	6.17 (5.92, 6.42)
Year slope (APC)	−0.78 (−1.16, −0.40)	−0.69 (−1.14, −0.24)	−0.63 (−1.17, −0.08)	−0.87 (−1.46, −0.27)
Output: categorical variables (percent change from base case)				
Tissue–base: fillet skin on				
Fillet skin off	5.0	NA	12.1	NA
Whole body	−13.7	NA	−34.0	NA
Waterbody type–base: lakes				
Great lakes	−57.8	−52.01	−28.1	−20.82
Impoundments	−5.4	NA	−15.9	NA
Rivers	−7.9	−10.71	−24.1	−10.56
Season–base: spring				
Fall	−17.0	−9.78	−17.9	−9.32
Summer	−8.3	−5.15	−4.2	−3.66
Winter	−9.8	−9.13	−15.2	0.300

Mixed model intercepts for MercNet were skin-on fillets in 1995 and for OMOE, skin-off fillets in 1990; also centered lengths differed among the species-datasets, *Pct/cm* percent change per cm increase in fish length, *APC* annual percent change

Largemouth bass from the Great Lakes were 28 and 21% lower for the respective datasets. These differences between the two datasets may be smaller than these values suggest given that the OMOE dataset combined lakes and impoundments. In MercNet, impoundments were 5 and 16% lower than lakes for walleye and largemouth bass. In rivers from MercNet, walleye Hg concentrations were 8% lower than in lakes and largemouth bass were 24% lower. In OMOE rivers, the two species were 11% lower than in lakes.

The northeast North American regional analysis of fish-Hg distributions (Kamman et al. 2005) evaluated Hg concentrations in 13 fish species and included a comparison among lakes, reservoirs, and rivers. Data since 1980 were used for spatial statistics, but temporal trends were not examined. In general, they found reservoirs had higher Hg

concentrations for larger fish species (including walleye), although this pattern was not seen for largemouth bass. Other previous statistical models of fish-Hg trends have not included the categorical variable of waterbody type. Previous studies have limited the Hg trend analysis to only lakes, excluding the river and impoundment data, because lakes are more likely to show a relationship to atmospheric deposition of Hg (e.g., Madsen and Stern 2007; Rasmussen et al. 2007; Monson 2009).

Seasonal differences were remarkably similar between species within datasets. The highest Hg concentrations were consistently in fish collected in the spring. In MercNet, fish collected in fall were 17–18% lower than those caught in spring; in OMOE, the fall collections were 9–10% lower. This seasonal difference concurs with analysis of the Wisconsin walleye subset by Rasmussen et al.

(2007). Higher Hg concentrations in the spring were likely attributable to changes in fish body condition, which has been shown to improve from spring to fall in Lake Erie walleye (Hartman and Margraf 2006), and is negatively correlated to fish-Hg concentration (Sun and Hitchin 1990; Greenfield et al. 2001; Dittman and Driscoll 2009).

Interaction terms were within one standard error of zero (not shown) and, therefore, did not significantly affect the modeled predictions.

Temporal trends

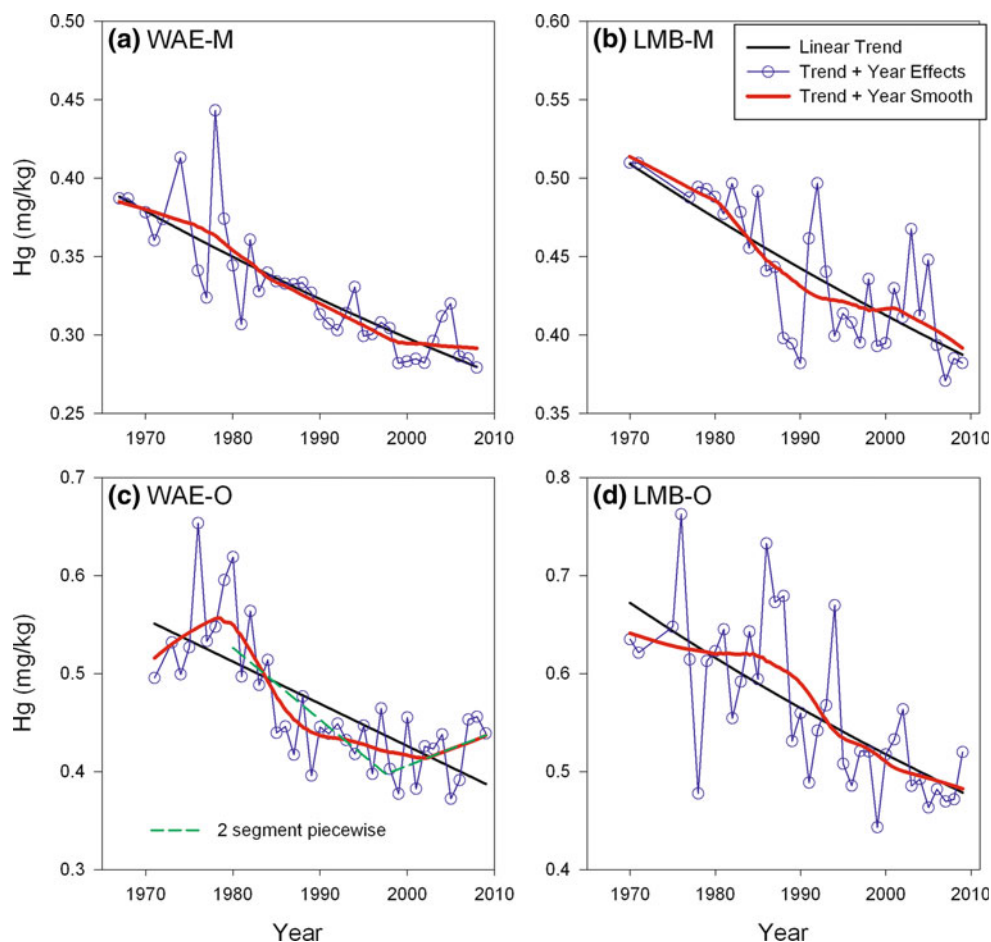
Deviations from the linear trend were highlighted by overlaying annual deviations from the linear trend on the linear trend line for each species-dataset (Fig. 3). In walleye from both datasets, Hg concentration trends appeared to level off beginning in the 1990s, and the Ontario walleye trend shifted upward in the last decade (Fig. 3a, c). This upward trend was statistically tested with a 2-segment linear piecewise regression for the period beginning in 1980. The regression algorithm found a significant breakpoint in 1997 (1997.5; S.E. 2.6; $P < 0.0001$) (Fig. 3c). Largemouth bass from both datasets continued the

downward trend in Hg concentration to the end of the period of record (Fig. 3b, d). Intercepts for largemouth bass were higher than those for walleye because all four sets were determined for a 42 cm fish length (see discussion above regarding differences between species at equal lengths).

The general downward trend in fish-Hg concentrations throughout the GLR was expected, given the reductions in Hg emissions from North America and Europe since the 1990s. Major Hg emission reductions occurred in the removal of Hg from latex paints, and controls on solid and medical waste combustion. However, two analyses of wet deposition data for Hg from the North American Mercury Deposition Network have not found a consistent temporal trend across the GLR (Butler et al. 2008; Prestbo and Gay 2009). Pacyna et al. (2006) estimated that worldwide emissions increased 16% between 1990 and 2000, and Streets et al. (2009) calculated an increase of 17% between 1996 and 2006. Trends in dry deposition of Hg are unknown.

The shift to an upward trend in OMOE walleye follows a pattern seen in Hg concentrations in fish from the Canadian Arctic, Greenland, and Lake Erie. Hg

Fig. 3 Linear temporal trend and random year deviations from the trend for each species dataset. Loess smooth of year deviations. Plot c includes a 2-segment piecewise linear regression from 1980 to end of record. **a** MercNet walleye. **b** MercNet largemouth bass. **c** OMOE walleye. **d** OMOE largemouth bass



concentrations in burbot (*Lota lota*) from the Mackenzie River (Fort Good Hope, Northwest Territories, Canada) increased by a factor of 1.6 between 1990 and 2008 (Carrie et al. 2010). Hg concentrations in Arctic char (*Salvelinus alpinus*) from a small lake in southwest Greenland increased 3.2% per year between 1994 and 2008 (Riget et al. 2010). Both of these studies attributed the Hg increase to warming climate. A comparison between 1996 and 2006 Hg concentrations in yellow perch from lakes within the maritime area of Kejimikujik National Park, Canada, showed that Hg in perch increased significantly in 10 of the 16 lakes studied (Wyn et al. 2010).

Subsets of the MercNet database have been previously analyzed and shown to have upward trends in some areas of the GLR. An analysis of fish-Hg concentrations in the Great Lakes demonstrated an increase in Hg concentrations in walleye from Lake Erie, although the trend was flat for walleye in Lake Ontario and declining for walleye in Lakes Superior and Huron (Bhavsar et al. 2010). Similar to the WAE-O pattern shown in our study, walleye and northern pike Hg concentrations in Minnesota lakes exhibited a biphasic pattern, with a downward trend between 1982 and

mid-1990s followed by an upward trend through 2006 (Monson 2009). In southern Wisconsin, walleye Hg concentrations increased at a rate of 0.6–0.8% between 1982 and 2005, while decreasing in northern Wisconsin between 1982 and 2005 (Madsen and Stern 2007; Rasmussen et al. 2007).

Spatial trends

The BLUPs for the random station-specific intercepts and slopes of $\log_e$ Hg versus time regression were plotted against latitude and longitude to show spatial trends in fish-Hg concentration and trends. Predicted site mean Hg concentrations are shown on a linear scale by plotting the antilog of the intercept BLUPs (Figs. 4, 5). Slope BLUPs were converted to APC by calculating $(e^{\beta_1 - \text{BLUP}} - 1) \times 100$ (Figs. 6, 7). For walleye, average predicted site means showed an increase with latitude, as indicated by a lowess smoother (red line) of the site means (Fig. 4a, c). Largemouth bass also showed an average increase with latitude (Fig. 4b, d), although the trend appeared more as a step function rather than a monotonic trend seen in walleye.

Fig. 4 Predicted site mean Hg concentrations (best linear unbiased predictors—BLUPs—of intercepts) by latitude for **a** MercNet walleye, **b** MercNet largemouth bass, **c** OMOE walleye, and **d** OMOE largemouth bass

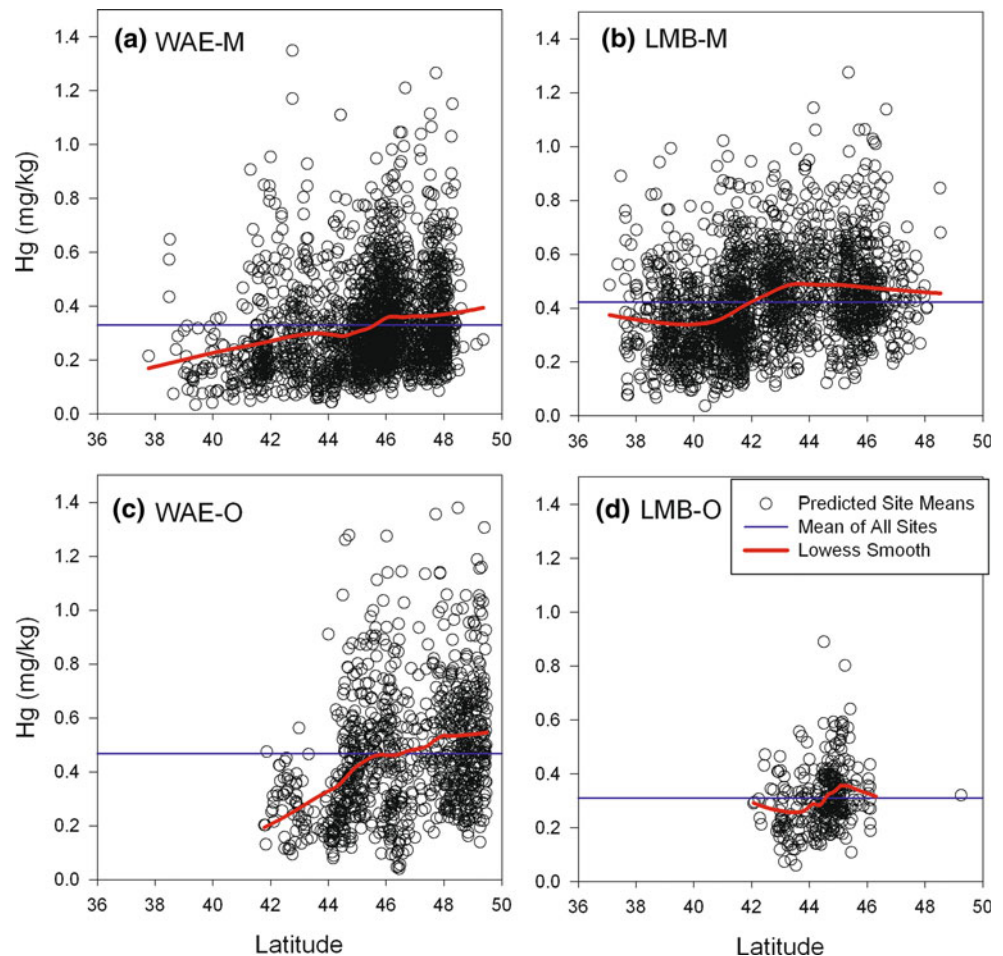
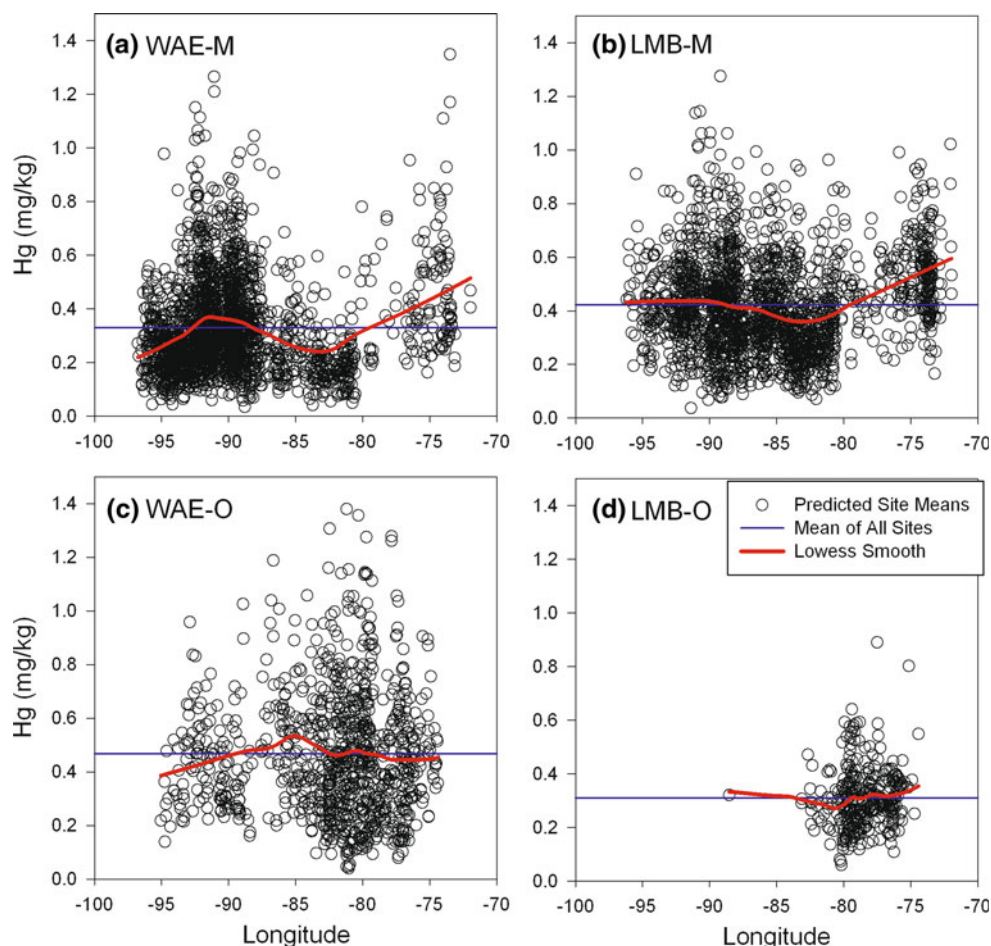


Fig. 5 Predicted site mean Hg concentrations (best linear unbiased predictors—BLUPs—of intercepts) by longitude for **a** MercNet walleye, **b** MercNet largemouth bass, **c** OMOE walleye, and **d** OMOE largemouth bass



The spatial trend in average predicted site means by longitude was not monotonic and differed between MercNet and OMOE. The MercNet species showed a similar pattern of increasing average site mean Hg concentrations in the eastern region (i.e., New York and Pennsylvania) (Fig. 5a, b). Average site means for OMOE walleye were relatively constant across the longitude range (Fig. 5c). OMOE largemouth bass were collected within a comparatively small longitudinal range (Fig. 5d), thereby restricting any observations of longitudinal trends.

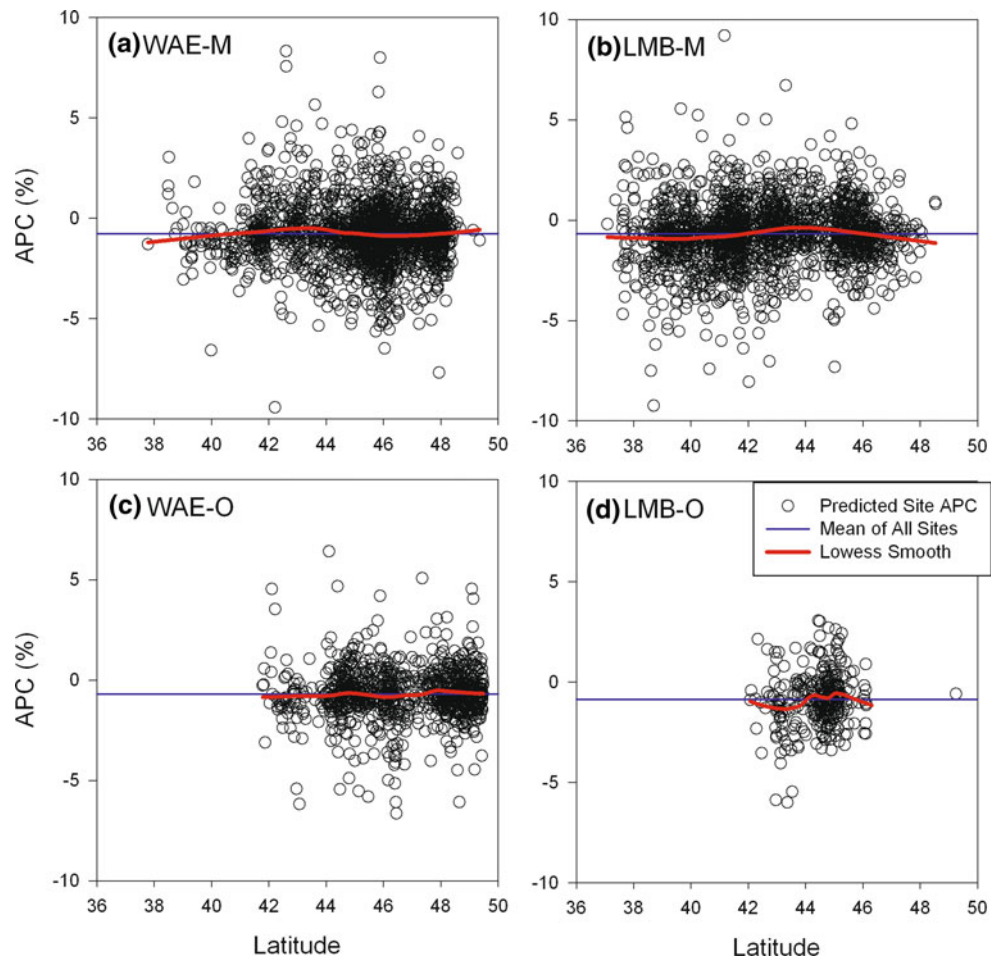
Unlike predicted site means, predicted site slopes did not show a spatial trend by latitude (Fig. 6). Although the overall average APC was about 1% for all four datasets, many sites were predicted to have positive trends (Fig. 6). Spatial trends of average APC by longitude, in contrast to latitude, showed an upward trend from west to east in MercNet walleye (Fig. 7a) and largemouth bass (Fig. 7c). Thus the spatial patterns with longitude were the same for predicted site means and site trends. The upward trend with longitude appeared to be driven by the higher and increasing Hg concentrations in the extreme eastern region.

Confounding factors

This observation of increasing Hg concentrations from west to east warrants subsequent investigation to assess the possible causes. Has there been increasing Hg deposition or increased mobilization of methylmercury in the eastern region; or is it a result of intensified monitoring of high Hg lakes in more recent years? Similarly, a logical next step in the analysis of Hg concentrations in GLR fish would be to explore why some sites showed increased Hg concentrations while others decreased in the same subregions of the GLR. This analysis showed that overall Hg concentrations in GLR walleye and largemouth bass declined over the 1970–2009 period at the rate of 0.6–0.9% per year; however, many sites showed increasing trends.

Fish age and gender likely contributed to unexplained variance in fish-Hg concentrations. Age and gender were not, however, consistently included in the GLR contaminant monitoring programs. Length is used as a surrogate for age. Gender was included in the OMOE dataset, although gender is usually not a consideration for fish-consumption advisories (the basis for many of the GLR monitoring

Fig. 6 Predicted site trend (best linear unbiased predictors—BLUPs—of slopes) by latitude, expressed as APC for **a** MercNet walleye, **b** MercNet largemouth bass, **c** OMOE walleye, and **d** OMOE largemouth bass



programs). Statistical models of Wisconsin walleye have shown that males have higher Hg concentrations than females of equal size (Madsen and Stern 2007; Rasmussen et al. 2007). A similar difference between male and female Ontario walleye was reported; however, other species, such as smallmouth bass, did not demonstrate a gender effect on Hg concentrations (Gewurtz et al. 2011). The difference in Hg concentrations between male and female walleye was most likely because of the lower growth efficiency in males (Henderson et al. 2003).

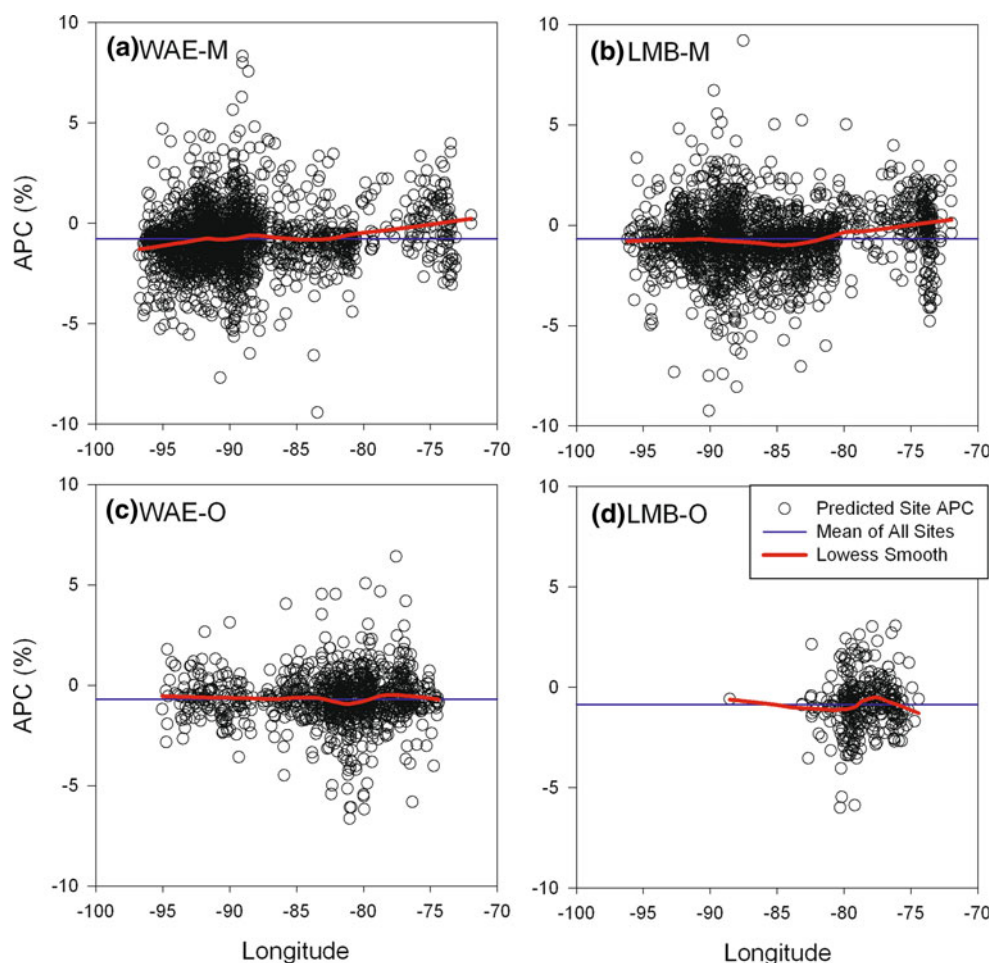
Differences in fish growth rate contribute to the variability of Hg concentrations, as well as increased fish-Hg concentrations with latitude. Many studies have reported lower fish-Hg concentrations in populations having higher growth rates (e.g., Harris and Bodaly 1998; Simoneau et al. 2005). Growth rates are negatively correlated to latitude in largemouth bass (Helser and Lai 2004) and walleye (Simoneau et al. 2005). Average water temperature is expected to decrease with latitude, and temperature affects fish growth rates (Jobling 1981). In an analysis of growth rates in more than 50 Quebec lakes, slower-growing walleye and northern pike had higher Hg concentrations at

a standardized length, and fish age at that length was positively correlated to latitude (Lavigne et al. 2010).

Fish growth rates are controlled by diet as well as temperature; therefore, changes to the food web will directly influence growth rate and Hg exposure. Aquatic invasive species have greatly affected food webs in the GLR. The invasive planktonic predator, spiny water flea *Bythotrephes longimanus*, has altered planktonic communities in water bodies throughout the GLR (Boudreau and Yan 2003; Chen and Folt 2005). Dreissenid mussels and the round goby (*Neogobius melanotomus*) invasions of the Great Lakes have created a new trophic pathway from sediment-bound Hg to benthic food webs (Hebert et al. 1989; Jude et al. 1992; Hogan et al. 2007).

These and many other factors will continue to complicate our ability to detect spatiotemporal trends in fish-Hg concentrations. Nevertheless, by accounting for some of the random and fixed effects, our study demonstrated that average Hg concentrations in two ubiquitous top predator fish in the GLR have trended downward between 1970 and 2009. This result supports the assertion that reductions in anthropogenic emissions of Hg are reflected

Fig. 7 Predicted site trend (best linear unbiased predictors—BLUPs—of slopes) by longitude, expressed as APC for **a** MercNet walleye, **b** MercNet largemouth bass, **c** OMOE walleye, and **d** OMOE largemouth bass



in the aquatic food web. Yet, the results also show deviations from a downward linear temporal trend in Ontario walleye and spatial trends of increasing Hg concentrations from south to north and west to east. Further analysis of spatiotemporal patterns should consider additional factors, such as temperature and growth efficiency. Confidence in the trends, and their causes, are expected to rise with the knowledge gained from extensive and persistent monitoring of fish-Hg concentrations and the relevant confounding factors (see Wiener et al. 2007).

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