

Temporal trends of mercury concentrations in Wisconsin walleye (*Sander vitreus*), 1982–2005

Paul W. Rasmussen · Candy S. Schrank ·
Patrick A. Campfield

Accepted: 26 June 2007 / Published online: 27 July 2007
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Abstract The Wisconsin Department of Natural Resources has monitored mercury (Hg) in several species of fish since the early 1970s primarily for fish consumption advisory purposes. We selected skin-on fillets of walleye (*Sander vitreus*) from inland lakes collected over the years 1982–2005 to assess temporal trends of Hg concentrations. While individual lakes are of interest, sample sizes, and unbalanced collections across fish lengths, seasons, or years prevent estimates of temporal trends of walleye Hg concentrations within most lakes. We evaluated temporal trends over all lakes using mixed effects models (3,024 records from 421 lakes). Relationships between Hg concentrations and a suite of lake chemistry, morphometry, and other variables were also explored. Hg concentrations generally increased with walleye length but the relationship varied among lakes. The best-fitting mixed effects models suggested that the overall rate of change in walleye Hg across all lakes in the dataset varied with latitude. Hg in walleye decreased 0.5% per year in northern lakes, increased 0.8% in southern lakes, and remained constant in middle latitude lakes over the period of 1982–2005. Season of collection was also an important predictor variable. Hg concentrations were highest in walleye captured in the spring and lowest in the fall. Other variables such as gender, lake area, and total alkalinity were also important predictors.

Keywords Mercury · Walleye · Temporal trends · Wisconsin · Bioaccumulation

Introduction

Mercury (Hg) concentrations in a variety of ecosystems have increased in the range of two to tenfold over preindustrial levels. Widespread Hg contamination and accumulation in fish poses health risks to people who consume fish and may impair wildlife health. Scientists have concluded that fish Hg concentrations will respond to changes in Hg loading rates. However, few reports demonstrate fish responses to control of Hg emissions and Hg inputs to surface waters from atmospheric loading. In addition, the magnitude and timing of the response of fish Hg to changes in atmospheric loading will vary due to a variety of factors influencing individual ecosystems (Munthe et al. 2007).

Most of the Hg that accumulates in fish is from anthropogenic sources and enters water via atmospheric deposition, accounting for most of the Hg in fish that inhabit remote lakes far from any point source (Glass and Sorensen 1999; Wiener et al. 2006). Sediment cores from lakes in Minnesota's Voyageurs National Park suggest Hg deposition rates increased after the mid-1800s with most Hg originating from anthropogenic sources (Wiener et al. 2006). Schuster et al. (2002) examined ice cores from the Fremont glacier in western Wyoming USA and found that Hg deposition increased during the industrial era, then decreased since the late 1980s possibly reflecting regulatory limitations on select sources of Hg to the environment. Estimated worldwide atmospheric Hg levels peaked in the late 1980s, decreased from the 1980s to 1996, and have varied little since 1996 (Slemr et al. 2003).

P. W. Rasmussen
Wisconsin Department of Natural Resources, Science Services,
2801 Progress Road, Madison, WI 53716-3339, USA

C. S. Schrank (✉) · P. A. Campfield
Wisconsin Department of Natural Resources,
Fisheries Management, 101 S. Webster St., Madison,
WI 53705-7921, USA
e-mail: Candy.Schrank@wisconsin.gov

Few studies have examined temporal trends in fish Hg from Hg-sensitive waters like the lakes typical of northern Wisconsin. Suns and Hitchin (1990) examined yearling yellow perch from 16 Canadian lakes and found no temporal trends in Hg concentrations over a 9-year period (1978–1987). The authors concluded that Hg inputs and conversion to bioavailable forms were also constant during this time. Hrabik and Watras (2002) found decreases in Hg deposition and Hg water column concentrations in one northern Wisconsin lake from 1994 to 2000. Hg concentrations in adult yellow perch from this lake also declined by 5% per year suggesting that Hg concentrations in adult fish decrease in response to reductions in Hg deposition. However, Hg trends in fish may not directly reflect changes in Hg emissions. Fish are exposed over a number of years and factors that affect bioaccumulation may vary spatially and temporally (Wiener et al. 2007).

Fish Hg concentrations increase with fish length, although the relationship varies among lakes (Lathrop et al. 1991; Somers and Jackson 1993). Some differences in Hg accumulation among lakes may be due to differences in fish growth rates (Simoneau et al. 2005). Studies have established the importance of lake morphometry and watershed characteristics in determining fish Hg levels (Bodaly et al. 1993; Rudd 1995; St. Louis et al. 1996). Lake chemistry, especially pH, alkalinity, and dissolved organic carbon, also affect Hg levels in fish (Wiener et al. 1990; Driscoll et al. 1995; Watras et al. 1998).

The Wisconsin Department of Natural Resources (WIDNR) has monitored Hg in fish since the early 1970s with the main purpose of determining appropriate advice for human consumption of fish. Monitoring designs evolved over time as more was learned about environmental contaminants and risks to human health from consumption of contaminated fish. Monitoring began in the 1970s to evaluate Hg concentration in fish from rivers receiving treated wastewater from facilities that used Hg-based slimicides. In the 1980s, atmospheric deposition, methylation and bioaccumulation of Hg in the food chain became a concern. When Wiener (1983) reported elevated Hg concentrations in sport fish from northern Wisconsin, the focus of monitoring shifted to lakes in that area. As a result, statewide Hg monitoring in Wisconsin fish was generally not conducted under a single study design that controlled spatial and temporal sampling patterns. Sampling for fish consumption advisories focuses on popular fishing lakes. However, the data also include fish collected for other purposes, including special studies of individual lakes, and general sport fish surveys. Each sample is affected by many uncontrolled factors, including lake characteristics such as water chemistry, underlying geology, watershed position, local climate, and local landscape cover, as well as temporal and spatial patterns of sampling.

Because of the large number of fish sampled and the 29-year time span, these data provide a valuable resource for examining spatial and temporal patterns of Hg contamination in Wisconsin fish.

Our objectives were to evaluate temporal trends while accounting for factors not controlled or randomized during sampling. We also wanted to characterize Wisconsin's walleye Hg concentrations in relation to fish length and lake attributes that have been shown to be important to Hg accumulation including lake morphometry, watershed, chemistry characteristics.

Methods

Description of datasets

Wisconsin Department of Natural Resource's fish contaminant database contains Hg results for 4,961 skin-on fillets from walleye collected over the years 1977–2005 from 607 locations. Following the recommendations of Wiener et al. (2007), we selected walleye skin-on fillets for our analysis because they are an important game fish highly sought by anglers, represent the largest number of observations in Wisconsin's fish contaminant database, and are a top-level piscivorous species that accumulate high concentrations of Hg. A dataset was selected for trend analysis by removing records from riverine-dominated flowages which may obscure responses to changes in atmospheric Hg loading (Wiener et al. 2007). WIDNR staff collected most walleye samples using typical fisheries survey methods, including electrofishing and netting (*Fisheries Management Handbook 3605.9, Contaminant Fish Collection Procedures, and the WIDNR Field Procedures Manual Version IV*). After collection and labeling, samples were frozen and later transported to a processing facility where they were measured, weighed, and filleted. Gender was determined for most mature individuals by gross examination of the gonads. Fish age was estimated for a subset of samples using scales and spines. Skin-on fillets were homogenized and subsamples placed in glass jars. Samples were analyzed for total Hg content according to the methods described in Sullivan and Delfino (1982). Total Hg in fish is comprised almost entirely of methylmercury (Weiner et al. 2007). Results were reported to two significant digits and the level of detection was 0.03 µg/g for samples analyzed prior to June 1994 and 0.004 µg/g after that date.

We compiled data on lake morphometry, hydrology, and water chemistry variables previously shown to affect Hg accumulation in fish from WIDNR databases for the lakes included in the walleye Hg trend dataset ($n = 421$). Water chemistry values were measured using conventional

laboratory procedures (Wisconsin State Laboratory of Hygiene 2005). We calculated lake-specific water chemistry values for statistical comparisons and modeling as the geometric mean of values from multiple years for each season, and then calculated a grand mean for each individual lake. Only results from surface samples were included in the calculations in order to standardize the data.

Statistical analyses

All statistical analyses were performed using SAS Version 9.1 (SAS Institute, Cary, NC, USA). We used log-transformed Hg concentrations ($\log_{10}(\text{Hg})$) as the response variable for analyses because the variance of residuals was more homogeneous on this scale. We converted year of collection to a value equal to the number of years since 1980. For example, a collection year of 1992 was given a value of 12 (1992–1980). Not all predictor variables were available for all lakes. Therefore, the number of lakes included in the analyses varied by the selected model. We log-transformed waterbody area and adjoining wetland area to linearize the relationship between $\log_{10}(\text{Hg})$ and those predictors. We approximately centered fish length and latitude predictor variables by subtracting overall mean length (45 cm) and latitude (45°), respectively, from each observation. Centering makes model intercepts more easily interpretable.

We analyzed log-transformed Hg concentrations using mixed effects models (Littell et al. 1996). Because this was an observational study, with sampling units selected by means outside of our control (Johnson 2002), and with many possible predictor variables, extra care was necessary in selection of predictor variables and specification of a set of candidate models to avoid overfitting and finding of spurious relationships (Burnham and Anderson 1998). Predictor variables were included in our models if we were specifically interested in estimating their effects, if we thought it was essential to adjust for their effects to obtain unbiased estimates of other parameters, or if they were shown to be important to Hg bioaccumulation. We included lake latitude, season in which each fish was collected, and fish length (cm) in our models to account for possible changes in the spatial distribution of lakes sampled, the seasonal timing of walleye sampling, and the size of walleye targeted for sampling. Lake latitude is also correlated with important gradients in pH and alkalinity, lake productivity, and fish growth (Colby and Nepszy 1981; Lillie and Mason 1983; Lathrop et al. 1989; Quist et al. 2003; Nate 2004). Fish length is known to be an important determinant of walleye Hg concentrations and must be accounted for in comparing lakes or estimating trends (Wiener et al. 1990; Lathrop et al. 1991). Water chemistry data were available for only a subset of the lakes

sampled for walleye Hg. We included total alkalinity in some of our models because it was available more often than other lake chemistry variables, is strongly correlated with other variables such as lake pH, and has been used in other analyses of Wisconsin walleye Hg data (Lathrop et al. 1991). We also examined the effects of some lake morphometry and watershed variables, including maximum depth, lake area, adjoining wetland area, and wetland to lake area ratio.

Both individual fish and whole lake predictor variables were included as fixed effects in our models. All models also included random effects to describe the variance of parameters among individual lakes. The random effects allow individual lakes to deviate from the population model determined by the fixed effects. The simplest model of this form allows lakes to deviate from the overall population intercept but not from the single population value for each of the coefficients (i.e., slopes) of the fixed factors. More complicated models for the random effects allow individual lakes to deviate from the population values of the coefficients for one or more of the fixed factors. We included random effects for the intercept, for fish length, and for year when selecting among models with different fixed effects structures (year was included as a random effect only for models in which it was also included as a fixed effect).

We fit models in two stages. We first fit a set of models that allowed us to estimate trends in walleye Hg, while accounting for factors that might bias trend estimates. We included models without year as a factor (i.e., a trend of zero) to provide a direct comparison for the same models with year. We ranked models on the basis of the Akaike Information Criterion (AIC) and examined models within 2 AIC units of the best-fitting model in greater detail, evaluating the random effects structure, and assessing model assumptions. We then fit additional models building on the best model from stage 1 by including factors related to lake morphometry, watershed characteristics, and gender. Model comparisons using AIC were based on maximum likelihood (ML) estimation, while final parameter estimates were obtained using restricted maximum likelihood (REML), as recommended by Verbeke and Molenberghs (2000).

Results

Data Summary

Selection of walleye, skin-on fillet records from inland, non-riverine lakes produced a dataset of 3,024 samples collected over the years 1982–2005 from 421 lakes. Individual samples represent a range of walleye size, age, and

Hg concentrations (Table 1). The bulk of the samples were collected between 1985 and 1994 (77% of records) with 2% from before 1985, and 21% from 1995 to 2005. Records were assigned to a season based on the month of sampling. The majority (73.7%) of the samples were collected in the spring (April–June) with 15.6% collected in summer (July–September) and 10.7% collected in fall (October–December). Gender was available for a subset of walleye samples ($n = 1953$). Of these 43.7% were females and 56.3% were males. The remaining were of uncertain gender either because the fish were not sexually mature or gender was not recorded.

The 421 lakes included in the dataset represent a range of hydrologic, morphometric, and water chemistry conditions (Table 2). The lakes are distributed in 49 of Wisconsin's 72 counties. A majority of the lakes (82%) are within the 20 most northern counties, approximating the distribution of inland lakes in Wisconsin. Hydrologic conditions include: drainage (58%) lakes that have both surface water inflow and outflow and stream drainage is the primary water source; seepage (30%) lakes that have no surface water inflow or outflow and precipitation or runoff is the primary water source; spring (8%) lakes that have no inlet and groundwater is the primary water source; and drained (3%) lakes that only have a surface water outflow and precipitation or runoff is the primary water source.

We evaluated the characteristics of the walleye records for all 421 lakes in the dataset over all years sampled (Table 3). Lake mean concentrations of Hg ranged 0.063–2.2 and the overall average was 0.483 $\mu\text{g/g}$. Lake mean fish length ranged from 31 to 69 cm. The years and number of years sampled varies by lake and the number of samples per lake ranged 1–36.

Models describing Hg bioaccumulation and trends

Mercury concentrations increased with walleye length, but the relationship varied among lakes (Fig. 1). Lake differences accounted for 54.7% of the total variation in walleye Hg values, while fish length explained 52.9% of the within lake variation in walleye Hg. Of the models without lake morphometry and watershed variables, the best fitting model included fish length, season, year, latitude, and the latitude by year interaction (Model 1, Table 4).

Including lake mean values for length of fish or year sampled improved the fit of the model slightly (~ 1 AIC unit), but did not change parameter estimates for the within-lake effects of length and year (i.e., there was no evidence for bias due to factors such as sampling changes related to fish length or year). This model included random effects for the intercept, fish length, and year; removing any one of these random effects from the model substantially reduced the fit of the model (AIC increased by 100 or more). These random effects allowed individual lakes to deviate from the overall population values for the intercept and for the slopes with respect to fish length and year sampled. The variability introduced by the random effects for the intercept and fish length is illustrated by plotting predicted Hg for all lakes in a given year (Fig. 2; predictions are for 1995). The variance estimates for the random effects of fish length and year sampled were small (Table 6), but their inclusion greatly improved the fit of the model.

The parameter estimates for the fixed effects model provide the overall population values for these parameters (Table 6). Because of random effects in the model, individual lakes deviated from population values for the intercept and the effects of fish length and year, but not for the effects of season, latitude, and the latitude by year interaction (latitude has only one value for each lake in any case). Although Hg was log-transformed prior to model fitting, parameter estimates are more interpretable when converted back to the original scale. Linear relationships on the log scale become exponential relationships on the original scale (Fig. 2); thus, the linear increase in $\log_{10}(\text{Hg})$ of 0.0181 per cm of fish length corresponds to a constant percent increase in Hg of 4.3% per cm $[(10^{0.0181} - 1) \times 100 = 4.3\%]$. Because fish length was centered at 45 cm and latitude was centered at 45° , with year measured as years since 1980, the intercept of -0.5277 corresponds to a Hg concentration of 0.30 $\mu\text{g/g}$ Hg ($10^{-0.5277}$) for a 45 cm fish from a lake at 45° north latitude caught in the summer of 1980. Seasonal effects were measured as deviations from summer values, with fish caught in the spring being 12.8% $[(10^{0.0597} - 1) \times 100]$ higher in Hg concentration than those caught in the summer, and fish caught in the fall being -1.5% $[(10^{-0.0064} - 1) \times 100]$ lower than summer-caught fish. Hg increased with latitude, but this relationship

Table 1 Descriptive statistics of walleye (skin-on fillets) records included in mercury trend analysis (WIDNR 1982–2005)

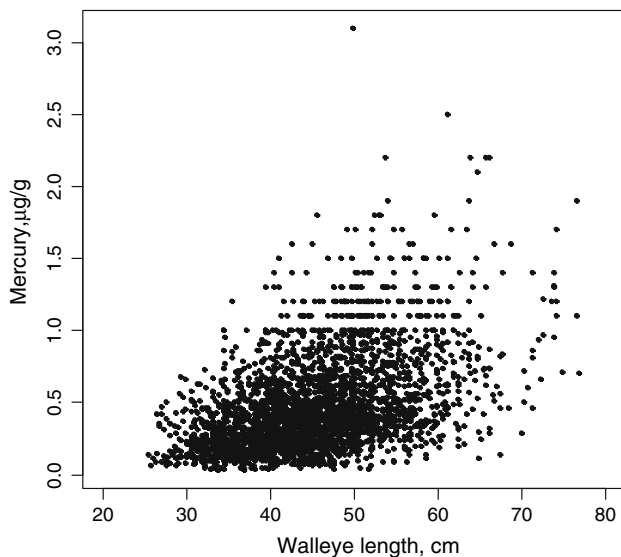
Variable	<i>n</i>	Mean	SD	C.V.	Minimum	Maximum
Fish length (cm)	3,024	45.0	8.5	19	25.4	76.7
Fish weight (kg)	2,986	0.94	0.64	68	0.1	5.46
Age (year)	160	5.5	2.1	39	2	12
Total mercury concentration ($\mu\text{g/g}$)	3,024	0.474	0.326	69	0.030	3.1

Table 2 Descriptive statistics of study lakes with selected mercury records

Variable	<i>n</i>	Mean	SD	C.V	Minimum	Maximum
Max depth (meters)	421	11.8	7.6	64	1.8	72.0
Drainage area (sq miles)	189	14	46	337	1	491
Watershed area (sq miles)	189	75	319	426	1	3,929
Waterbody size (acres)	421	1,045	6,818	652	17	137,708
Wetland adjoin area (acres)	421	541	3,932	727	0	78,060
Wetland-Lk area ratio	421	0.91	5.39	591	0	87.89
pH (geo. mean, standard units)	137	7.9	0.6	8	5.7	9.5
Alkalinity (geo. mean, mg/l total CaCO ₃)	204	63.7	54.4	85	2.8	213
Color (geo. mean, standard units)	207	27	28	105	5	150
Latitude (decimal degrees)	421	45.5196	0.8286	2	42.5916	46.7951
Longitude (decimal degrees)	421	-90.0611	1.1787	1	-92.711	-87.1542

Table 3 Descriptive statistics of lake means of walleye (skin-on fillet) records included in this study

Variable	<i>n</i>	Mean	SD	C.V.	Minimum	Maximum
Lake mean Hg concentration (µg/g)	421	0.483	0.306	63	0.063	2.2
Lake mean fish length (cm)	421	46.00	5.89	13	30.73	69.34
Lake mean year sampled	421	1990.5	4.8	0.2	1983.7	2004.0
Lake mean number of years sampled	421	1.5	0.9	57	1.0	7.0
Lake mean number of mercury results	421	7	6	80	1	36

**Fig. 1** Observed total Hg concentration (µg/g) for 3,024 walleye skin-on fillet samples from 421 Wisconsin lakes

varied among years, as indicated by the interaction between latitude and years (Fig. 3). The rate of increase in Hg with respect to latitude was greater early in the sampling period than during recent years. Equivalently, Hg decreased in northern Wisconsin lakes during the sampling period, while Hg increased in southern lakes. Because of the exponential increase of Hg with fish length, these

relationships appear stronger for larger fish (Fig. 3). We confirmed these relationships by estimating time trends separately for southern, central, and northern Wisconsin lakes—trends in Hg concentrations (µg/g) averaged +0.8% per year (linear trend = 0.0034, SE = 0.0037) for southern lakes, were approximately zero (linear trend = -0.0002, SE = 0.0019) for central lakes, and averaged -0.5% per year (linear trend = -0.002, SE = 0.0028) for northern lakes.

Models including lake morphometry and watershed variables

Adding log of lake area or log of adjoining wetland area to Model 1 improved the fit; in particular, the model with log of lake area fit far better than any other model (Model 11, Table 5). Log of Hg concentration decreased as the log of lake area increased and the variance among lake intercepts was reduced from its value in the simpler model because this model accounted for more of the lake differences (Table 5). Because both Hg concentration and lake area were log-transformed, the slope of -0.1487 indicates that if lake size is doubled, Hg concentration decreases by 9.8%.

Models including lake alkalinity and gender of fish

Because alkalinity was not available for all lakes and gender not known for all fish, we used subsets of the data to

Table 4 Models fit by maximum likelihood (*ML*), listed in order with best fit first

Model number	Fixed effects	Δ AIC
Model 1	L Y S lat lat*Y	0
Model 2	L Y S lat	4.6
Model 3	L Y S	21.3
Model 4	L Y lat lat*Y	25.6
Model 5	L Y lat	31.4
Model 6	L Y	53.2
Model 7	L S lat	110.7
Model 8	L S	125.5
Model 9	L lat	177.7
Model 10	L	198.4

Δ AIC is the difference in AIC from best-fitting model (model with minimum AIC). All models include an intercept. Abbreviations used for factors in models are: *L* fish length, *lat* latitude, *S* season, *Y* year sampled, *lat*Y* latitude by year interaction

Table 5 Models based on best model from Table 4, but also including lake morphometry and watershed data

Model number	Fixed effects	Δ AIC
Model 11	L Y S lat lat*Y logArea	0
Model 12	L Y S lat lat*Y logWetland	38.1
Model 13	L Y S lat lat*Y	42.0
Model 14	L Y S lat lat*Y ratio	43.6
Model 15	L Y S lat lat*Y depth	43.8

Other variables included are: *logArea* log of waterbody area, *logWetland* log of adjoining wetland area, *depth* waterbody maximum depth, *ratio* ratio of adjoining wetland area to waterbody area

examine models including these predictor variables. Gender was known for 1,953 fish from 318 lakes. Alkalinity was available for 204 lakes, with 1,642 fish sampled. For each of these two subsets, we first fit the best model identified for all lakes (Model 11 with predictor variables fish length, season collected, year collected, lake latitude, year by latitude interaction, and log of lake area). Adding gender as a predictor variable significantly improved the fit of the model (AIC decreased by 68.8). Walleye Hg was 14.7% lower $[(10^{-0.069}-1) \times 100]$ for females than for males of equal size. Adding log of alkalinity as a predictor variable significantly improved the fit of the model (AIC decreased by 23.8). Walleye Hg decreased by 14.4% as alkalinity doubled. The model including fish length, season collected, year collected, latitude, the latitude by year interaction, log of lake area, and the log of alkalinity fit approximately as well as a slightly simpler model that did not include latitude or the latitude by year interaction, but included instead the interaction between year and log of alkalinity (AIC for the latter model was 0.5 smaller). This suggests that the geographical pattern in walleye Hg time

trends may be related to water chemistry differences between northern and southern lakes. In fact, in our data set latitude and alkalinity were strongly negatively correlated ($r = -0.82$, $P < 0.001$).

Discussion

Our analyses suggest that temporal trends in walleye Hg concentrations vary latitudinally within WI, with northern lakes exhibiting slight average decreases (-0.5% per year) and southern lakes modest average increases in Hg concentration (0.8% per year) over the time period from 1982 to 2005. The random effects associated with time trends indicate that individual lakes may deviate from these population averages. For the subset of lakes on which alkalinity was measured, we found that models in which time trend differences were related to alkalinity or to latitude fit equally well. Because northern Wisconsin lakes are generally lower in alkalinity and southern lakes generally higher, it is difficult to separate the effects of alkalinity and latitude in our observational study. Furthermore, other factors, including lake chemistry, atmospheric deposition, lake productivity, and fish growth rates, are also associated with latitude and alkalinity, so that several explanations for patterns in time trends are possible. Regional and local differences in walleye Hg trends are likely because of the complex interplay between atmospheric deposition of Hg, lake alkalinity and pH, and factors that affect microbial community composition and activity, such as the nutrient loading from runoff, or climate change (Watras et al. 2006).

Our finding that walleye Hg concentrations decreased in northern Wisconsin is consistent with other studies. Hg concentrations in northern pike and walleye from Minnesota declined by an average of 11% between 1990 and 2000, or $\sim 1\%$ per year (MPCA 2007). Madsen and Stern (2007) found walleyes (skin-on and skin-off fillets) from northern Wisconsin decreased 0.6%/year over 1982–2005 using hierarchical Bayesian methods. In a study of common loons in northern Wisconsin, Fevold et al. (2003) estimated that adult loon blood Hg levels decreased by 4.2% per year, while loon chick blood Hg decreased by 4.9% per year. Engstrom and Swain (1997) found sediment cores showed evidence of recent declines in Hg deposition to lakes in northeastern Minnesota after reaching a peak in the 1960s–1970s. Hrabik and Watras (2002) attributed the decreases in yellow perch Hg concentration to decreases in atmospheric deposition of H_2SO_4 and Hg that occurred in northern Wisconsin after 1988 (Watras et al. 2000). Sulfate deposition and frequency of acidic precipitation has decreased throughout the Midwest (Lehman et al. 2007). In the poorly buffered lakes typical of northern Wisconsin,

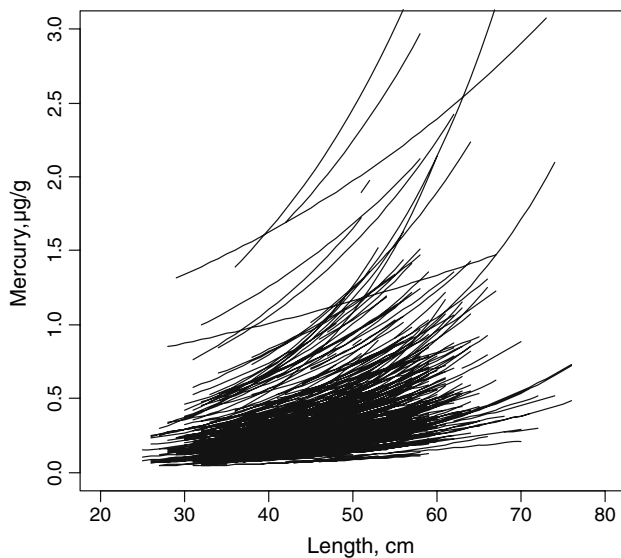


Fig. 2 Predicted total Hg for the year 1995 from best fitting model (Model 1) without variables on lake morphometry or watershed data. Each line represents predictions for fish from one lake, over the size range of fish observed for that lake

lakes. Also, the well buffered lakes of southern Wisconsin are much less likely to show changes in lake chemistry due to regional decrease in sulfate deposition than are the poorly buffered lakes of northern Wisconsin. However, our finding of increased walleye Hg in southern lakes warrants further study. There are fewer lakes in southern Wisconsin and our data set includes fewer fish from southern lakes (573 fish from 62 lakes south of 45° latitude).

Researchers have long recognized that mean walleye Hg concentrations vary greatly among lakes, and have sought to find factors to explain this (e.g., Wiener et al. 1990; Lathrop et al. 1991; Wren et al. 1991). In our analyses, lake differences were expressed as differences in both the intercept and slope of the $\log_{10}(\text{Hg})$ —walleye length relationship. Although our study was not directed primarily at examining factors that account for lake differences in walleye Hg concentration, we found that lake latitude, area, and alkalinity explained some of the differences in intercepts among lakes, but that none accounted for slope differences. Neither lake latitude nor alkalinity are likely to be direct causes of differences in walleye Hg concentrations,

Table 6 Parameter estimates from best models (model 1 and 11) fit by restricted maximum likelihood (REML)

Parameter	Model 1 estimate	SE	Model 11 estimate	SE
Intercept	-0.5277	0.0244	-0.1426	0.0618
Fish length	0.0181	0.0005	0.0180	0.0005
Latitude	0.1072	0.0227	0.0950	0.0220
Year	0.0008	0.0016	0.0010	0.0015
Latitude*year	-0.0038	0.0015	-0.0038	0.0014
Log waterbody area	–	–	-0.1487	0.0220
Spring–summer	0.0597	0.0136	0.0620	0.0135
Fall–summer	-0.0064	0.0204	-0.0051	0.0202
<i>Variance components (expressed as standard deviations)</i>				
Intercept	0.2693		0.2574	
Fish length	0.0063		0.0063	
Year	0.0130		0.0127	
Within lake (residual)	0.1339		0.1340	

SE standard errors

decreases in sulfate deposition may change lake chemistry in ways that reduce the efficiency of Hg methylation (MPCA 2006; Munthe et al. 2007).

Our analyses also suggest slight increases in walleye Hg concentrations in southern Wisconsin. Hg deposition tends to be greater in southeastern Wisconsin and these lakes may be receiving more Hg than northern lakes. However, we do not have temporal trend estimates for Hg deposition to most Wisconsin lakes (WIDNR 1999). Engstrom and Swain (1997) found no evidence of decreased Hg deposition in sediment core samples from west-central Minnesota

but both are probably correlated with other causative factors. We included latitude in our models, not only to account for spatial sampling patterns in our observational study, but also because it is generally associated with gradients in pH and alkalinity, lake productivity, and fish growth, all factors suspected or demonstrated to be important to Hg bioaccumulation. In Wisconsin, pH and alkalinity, lake productivity, and fish growth rates all tend to decrease with increasing latitude (Lillie and Mason 1983; Nate 2004). Alkalinity and pH themselves are probably correlated with causative factors such as the

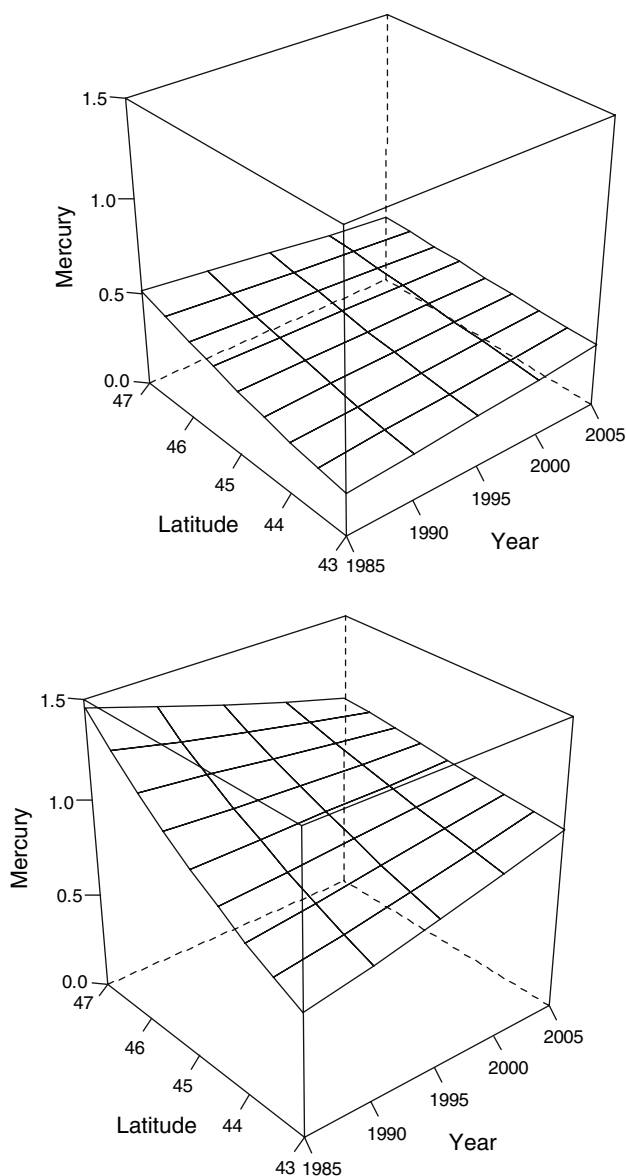


Fig. 3 Predicted mercury ($\mu\text{g/g}$) in skin-on fillets of a 45 cm walleye (*top panel*) and a 70 cm walleye (*bottom panel*) by year sampled and lake latitude (decimal degrees)

activity of sulfate-reducing bacteria leading to increased rates of methyl Hg production (Watras et al. 2006). Increased lake productivity and warmer water temperatures in southern Wisconsin lead to faster fish growth rates, which Simoneau et al. (2005) have found to be associated with lower Hg concentrations in fish of the same length. High lake productivity is also associated with higher densities of phytoplankton and zooplankton and reduced rates of Hg trophic transfer (Chen and Folt 2005). Both Bodaly et al. (1993) and Greenfield et al. (2001) found that lake size and fish Hg are negatively related, just as we did. They suggested that this was due either to higher temperatures in

smaller lakes, leading to higher rates of methylation, or to proportional increases in organic matter input in smaller lakes, which tends to reduce oxygen levels and increase rates of methylation by sulfate-reducing bacteria.

Differences in walleye Hg among fish within lakes were related to fish length, gender, and the season of collection. The increase in Hg concentration with fish length is well known (MacCrimmon et al. 1983; Wiener et al. 1990; Wiener and Spry 1996) and has been modeled using mechanistic models that account for bioenergetic and physiological parameters (Trudel and Rasmussen 2001). The higher concentration of Hg in males than females at the same length is probably due to the higher growth rate of females; males take longer to reach the same size, and consume more Hg during that time (Henderson et al. 2003). We found that for walleye of the same length, Hg concentrations were highest in fish collected in spring; this was also the season in which most of our samples were collected. Post et al. (1996) suggested that seasonal changes in Hg concentrations in fish are caused by seasonally varying diet and growth efficiency, although they found highest Hg concentrations in late summer for age-0 yellow perch. Fish Hg may also reflect seasonally varying methylmercury water column concentrations (Monson and Brezonik 1998; Eckley and Hintelmann 2006).

Our findings suggest that Hg in walleye from Wisconsin lakes changed in the range of -0.5 to 0.8% per year depending on geographical position in the state during the period of 1982–2005. These trends may reflect geographically differing temporal trends in the amount of Hg deposited to Wisconsin lakes. However, long-term changes in other factors, such as water chemistry, fish growth rates, and lake levels, known to impact Hg bioavailability and accumulation may also be important. These findings should be re-evaluated in future years.

Acknowledgments Special thanks to Drs. Michael W. Meyer and Nancy A. Nate for reviewing early drafts of this manuscript. Assistance of staff at the Wisconsin State Laboratory of Hygiene is appreciated. The Wisconsin Department of Natural Resources generated the data summarized in this report through efforts over many years supported by different sources of funding including state and federal programs. Collection of 2003–2005 samples was partially funded by the FOCUS on ENERGY Program administered by the Department of Administration. Data compilation and analysis was funded in part by the FOCUS on ENERGY Program.

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