

Mercury Concentrations in Lentic Fish Populations Related to Ecosystem and Watershed Characteristics

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Abstract Predicting mercury (Hg) concentrations of fishes at large spatial scales is a fundamental environmental challenge with the potential to improve human health. In this study, mercury concentrations were examined for five species across 161 lakes and ecosystem, and watershed parameters were investigated as explanatory variables in statistical models. For all species, Hg concentrations were significantly, positively related to wetland coverage. For three species (largemouth bass, pike, and walleye), Hg concentrations were significantly, negatively related to lake trophic state index (TSI), suggestive of growth biodilution. There were no significant relationships between ecosystem size and mercury concentrations. However, Hg concentrations were strongly, positively related to ecosystem size across species. Scores of small or remote lakes that have never been tested could be prioritized for testing using models akin to those presented in this article. Such an approach could also be useful for exploring how Hg concentrations of fishes might respond to natural or anthropogenic changes to ecosystems over time.

Keywords Contaminant · Ecology · Fish tissue · Food chain · GIS · Methylation · Wisconsin

Introduction

Human health is jeopardized if mercury (Hg) contaminated fishes are regularly consumed (Fitzgerald and Clarkson

1991; Mergler et al. 2007). Yet Hg concentrations in fishes can be extremely variable across ecosystems (Björklund et al. 1984; Drevnick et al. 2007; Lindeberg et al. 2007) making fish tissue monitoring a major challenge. Regulatory agencies in water-rich areas are often charged with the task of monitoring Hg in fishes from thousands of waterbodies, many of which have never been previously tested. Fish tissue monitoring programs could be enhanced with improved capacity to identify and anticipate problematic waterbodies and prioritize fish tissue testing so that consumption advisories can be issued more efficiently and with greater precision.

Certain ecosystem and watershed variables are already known or suspected to influence the production and accumulation of methylmercury (MeHg—i.e., the form that readily bioaccumulates) by aquatic organisms (Bonzongo and Lyons 2004; Warner et al. 2005; Belger and Forsberg 2006; Rypel et al. 2008). The primary sources of MeHg to lakes are the atmosphere, runoff from terrestrial catchments, and in-system production (St. Louis et al. 1994; Rudd 1995); more specifically, microbially catalyzed MeHg production in anaerobic bottom sediments (Warner et al. 2003). The relative importance of these sources varies across ecosystems, however, MeHg concentrations are often higher in ecosystems with environmental features that promote higher MeHg production/accumulation (Bonzongo and Lyons 2004; Driscoll et al. 2007). For example, wetlands are a known source of MeHg production and export to aquatic food webs, and ecosystems with large wetlands usually have heightened Hg concentrations in organisms (Warner et al. 2005; Rypel et al. 2008; St. Louis et al. 1994).

Likewise, ecosystem productivity can augment the availability, and thus the rate of MeHg accumulation by organisms through a process known as “growth

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biodilution”—i.e., how biomass-specific concentrations of metals diminish as cells divide (Pickhardt et al. 2002; Chen and Folt 2005). Growth biodilution is the product of one or several biologic scenarios (Chen and Folt 2005), but is a process ultimately driven by nutrient dynamics, primary production, and lake trophic state (i.e., ecosystem-level characteristics). This creates the interesting pattern of negative correlations between lake phytoplankton density and Hg burdens in phytoplankton and their consumers, e.g., fishes (Chen and Folt 2005).

Furthermore, ecosystem size (a variable historically studied in community ecology) has been implicated in unsafe Hg concentrations (Post et al. 2000; Post 2002). The idea is well developed that food chain length scales positively with ecosystem size (Post et al. 2000), and the logical extension of this idea is that contaminant concentrations might also scale positively with ecosystem size. However, this idea has never been tested. The objectives of this article were to simultaneously examine the relationships between wetland coverage, ecosystem productivity, ecosystem size, and Hg concentrations of fishes in Wisconsin lakes and to evaluate whether any of these potential relationships would be useful for managers.

Materials and Methods

All available mercury fish tissue data for Wisconsin USA lakes were retrieved for five fish species (Lake $N = 161$, Tissue sample $N = 1648$) from the United States Environmental Protection Agency (US EPA) national mercury database (<http://www.epa.gov/waterscience/fish/mercurydata.html>). Data were based on composite tissue samples field-collected and analyzed by US EPA, 1989–1995 using standard methods. Following Shapiro–Wilk tests (all P 's < 0.10), all mercury data were \log_{10} -transformed (+1) to meet assumptions of normality.

Hydrologically connected wetlands within watersheds were delineated by analysis of high-altitude aerial photographs using Google Earth Pro and consulting Wisconsin Wetland Inventory maps (<http://dnrm.wisconsin.gov>). Lake areas (i.e., ecosystem size) were delineated using the same aerial photographs and software. Wetland area was converted to a wetland area index (ranging from 0 to 100%) by dividing the area of wetlands hydrologically connected to each lake by the area of the lake plus wetlands, multiplied by 100. A trophic state index (TSI) value was obtained for each lake from the University of Wisconsin Lake Water Clarity Monitoring and Analysis database (www.lakesat.org). Low values indicate more oligotrophic lakes, whereas high values indicate more eutrophic conditions. TSI values were based on a 3-year composite of each lake's water clarity as it appeared on

satellite images taken on a late summer day in 1999, 2000, and 2001 calibrated against field Secchi or chlorophyll α measurements.

Forward stepwise multiple linear regressions (cutoff value for model entry, $\alpha < 0.05$) were calculated for each species using $\log + 1$ fish mercury concentrations as the dependant variable, and fish size (mean total length, mm), wetland coverage (%), TSI (%), and ecosystem size (area, km^2) as independent variables. Independent variables were considered to have a significant effect on fish Hg concentrations only if their beta value and model P were both significant at $\alpha < 0.05$. Using correlation matrices, it was found that no independent variables examined were significantly correlated with another and in no case were tolerance values of independent variables in regressions (the inverse of the variance inflation factor) > 1.0 . The complete range of environmental variables examined for lakes in this study can be found in Table 1. Furthermore, while some previous studies have extracted length effects prior to examining effects due to environmental factors, this approach was not used here. In this study, final models included length effects so that models would ultimately be more useful to managers (e.g., length effects for a given waterbody might be predictable in combination with knowledge of ecosystem and landscape factors). However, to illustrate Hg effects due to ecosystem and landscape factors in this paper (i.e., Fig. 1, panels a–c), length effects were removed by regression of Hg concentrations against fish length, and the residuals from these regression plotted against the environmental variables of interest (see Fig. 1, panels a–c). To examine whether Hg concentrations of fish scaled with ecosystem size across species, a linear regression was generated from the geometric means of $\log_{10}\text{Hg} + 1$ concentrations for each species against mean ecosystem size for each species [calculated from data in Wisconsin Department of Natural Resource DNR (Wisconsin Department of Natural Resources 2005)].

Results

Fish length significantly influenced Hg concentrations for all species (Table 2). Hg concentrations of all species were significantly and positively affected by the wetland area index (Table 2), however, the effect for bluegill was noticeably different from the other species examined (Fig. 1a). Hg concentrations of three species were significantly, negatively affected by TSI (Table 2; Fig. 1b). Ecosystem size did not affect the Hg concentrations of any species examined; however, directional effects were consistently negative (Table 2; Fig. 1c). A significant positive regression was generated relating mean Hg concentrations among species to mean ecosystem size ($P = 0.01$, $r^2 = 0.83$; Fig. 1d).

Table 1 Characteristics of lake ecosystems examined in this study

Species	Number of lakes	Fish size (TL, mm)		Ecosystem size (km ²)		TSI (%)		Wetland area index (%)		Hg (mg kg ⁻¹)	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
<i>Lepomis macrochirus</i>	47	181	130–227	2.24	0.07–20.39	47.2	33–72	35.5	1.4–89.9	0.120	0.048–0.255
<i>Micropterus salmoides</i>	66	351	279–498	1.71	0.07–20.39	47.4	33–72	35.7	1.6–89.9	0.364	0.062–0.765
<i>Esox lucius</i>	61	545	386–886	2.08	0.17–20.39	47.4	33–61	32.8	1.6–71.2	0.326	0.080–0.708
<i>Sander vitreus</i>	111	457	301–803	3.19	0.30–20.39	46.5	32–72	30.6	1.6–78.1	0.474	0.095–1.450
<i>Esox masquinongy</i>	32	854	408–1371	40.28	0.12–20.39	47.9	38–61	29.0	0.4–92.5	0.845	0.041–2.200

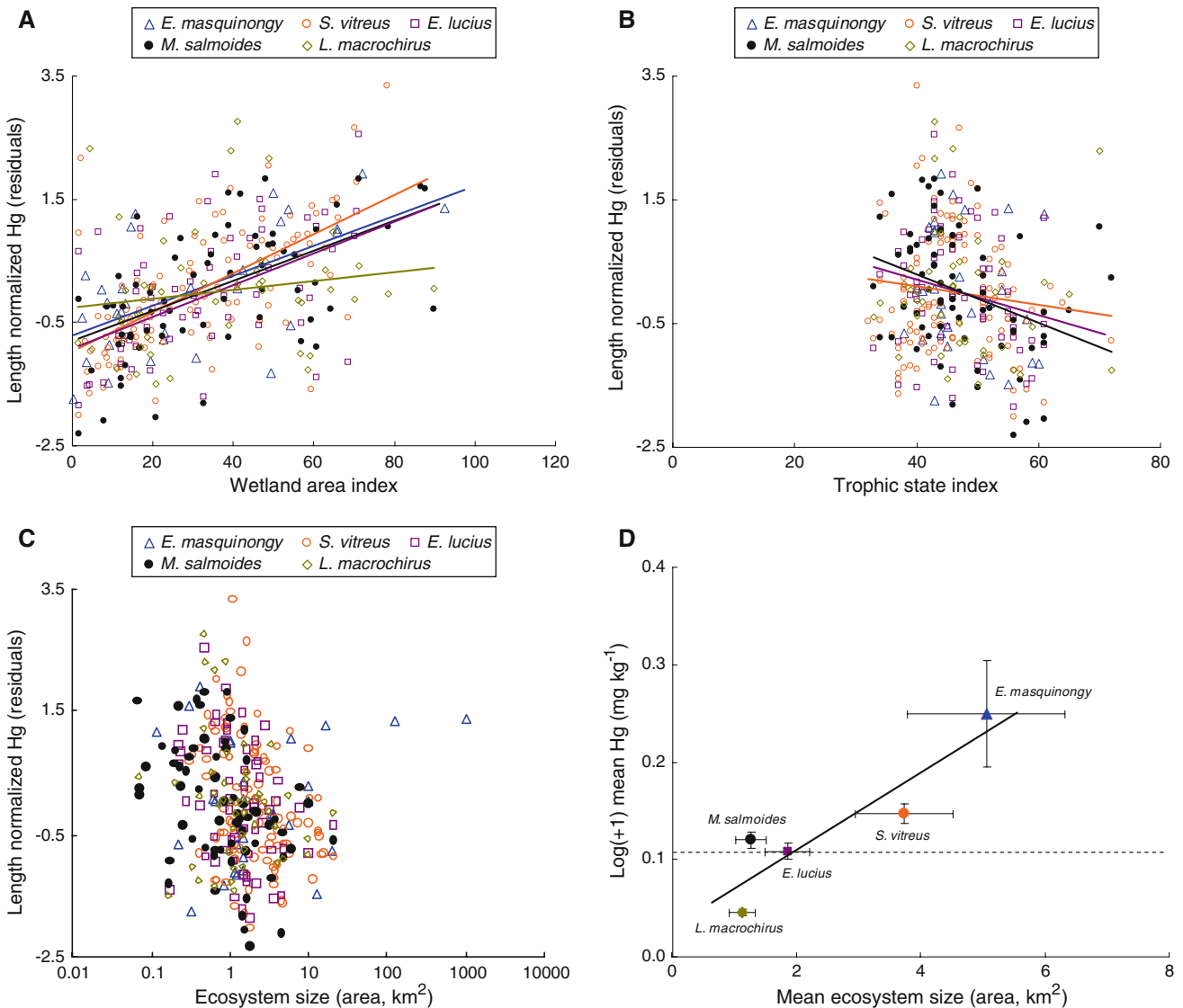


Fig. 1 Mercury concentrations of five fish species in Wisconsin lakes plotted against **a** wetland area index, **b** trophic state index, **c** ecosystem (lake) size, and **d** mean ecosystem size inhabited by each species ($\text{Log}(\text{Hg} + 1) = 0.04x + 0.03$, $R^2 = 0.83$), error bars represent the mean ± 1 SE). For panels **a**–**c**, Hg values were adjusted for effects of fish length by using standardized residual values from a fish

length-Hg regression. For panel **d**, mean Hg values ($\text{log} + 1$) represent geometric means. Horizontal dashed line represents the US EPA 0.3 mg kg⁻¹ action level for a human consumption advisory. Only significant factors in multiple linear regressions (from Table 2) are shown as regression lines in these panels

Discussion

If it is known (for numerous ecosystems in a region) how mercury concentrations of fish vary with ecosystem and watershed characteristics, species, food chain length, and fish size, then concentrations of Hg in fish from other ecosystems can be reasonably projected. Like other statistical models, results could potentially be improved with additional data or more variables of interest. The most desirable variables to include in the context of predicting fish Hg concentrations would be direct measures of microbial Hg methylation rate (Jeremiason et al. 2006) or field measurements of water chemistry such as pH, nutrients, dissolved oxygen (Qian et al. 2001). Yet, these data are simply not available for the droves of small or remote lakes for which fish Hg data are also lacking. In this study, the independent variables examined were available for every body of water studied, and can all be remotely generated (i.e., through analysis of Landsat images). Future models could potentially be supplemented by adding landscape factors (that are either widely available or capable of being remotely generated) that might explain additional variation in fish Hg content such as soil depth, soil type (e.g., organic carbon content), hydrologic flow regime, vegetation cover, and human land use classifications.

This approach (or one that is similar), can be applied to improve the efficacy of regulatory agencies at detecting waterbodies prone to dangerous Hg concentrations in fish. More specifically, the models can isolate which waterbodies possess the typical combination of wetland coverage, ecosystem productivity, and fish species that make them vulnerable to elevated Hg concentrations. Such an approach would be invaluable to prioritizing future fish tissue

monitoring efforts so that the ecosystems most likely to harbor dangerous Hg concentrations in fishes can be rapidly identified, tested, and human consumption advisories published as soon as possible.

However, this form of approach also holds potential for the equally important, but more challenging task, of addressing how fish mercury concentrations are changing over time in response to other important environmental drivers (e.g., changes in atmospheric mercury deposition, wetland reductions and expansions, invasive species, and climate change). Again, if it is understood how fish Hg concentrations correlate to variations in ecosystem and watershed characteristics, and it is then revealed how an environmental perturbation (e.g., Schindler and Smol 2006; Munthe et al. 2007) modifies these key variables or is changing through time, Hg concentrations in fishes can be modeled as ecosystems change. For example, additions and losses in the size of wetlands due to changes in wetland mitigation practices (Stokstad 2008), or ecosystem fragmentation (Rypel and Layman 2008) might reconfigure the dynamics of the mercury cycle in many systems. To what degree could this influence Hg levels in certain fishes and ecosystems? Models such as the ones presented here may be capable of exploring (at least initially) the potential outcomes of such change. However, the capacity to predict Hg concentrations in fish populations in this study was derived from several relationships that would also need to be apparent (or be based on new or differing relationships) in any future efforts.

First, Hg concentrations of fishes in lakes scaled significantly and positively with lake wetland coverage for all species. This was the strongest predictor of Hg concentrations for all but one species and was a better predictor of Hg concentrations than even fish size. Wetlands are a well-known source of MeHg to aquatic ecosystems. St. Louis

Table 2 Results of multiple regressions relating mercury concentrations for five species to fish length and ecosystem and watershed variables for Wisconsin lakes

Species	Length (<i>P</i> -value)	Wetland area index (<i>P</i> -value)	TSI (<i>P</i> -value)	Ecosystem size (<i>P</i> -value)	Model (<i>P</i> -value)	Model <i>R</i> ²	Model equation
<i>Lepomis macrochirus</i>	0.008 (+)	0.01 (+)	0.25 (–)	0.37 (–)	0.01	0.26	Log(Hg + 1) = –0.00929 + 0.00035 × Length – 0.00033 × TSI + 0.0003 × wetland area index
<i>Micropterus salmoides</i>	<0.0001 (+)	<0.0001 (+)	<0.0001 (–)	0.16 (–)	<0.0001	0.55	Log(Hg + 1) = –0.00321 + 0.00062 × Length – 0.00213 × TSI + 0.0013 × wetland area index
<i>Esox lucius</i>	0.007 (+)	<0.0001 (+)	0.04 (–)	0.38 (–)	<0.0001	0.34	Log(Hg + 1) = 0.054 + 0.00018 × Length – 0.0015 × TSI + 0.0012 × wetland area index
<i>Sander vitreus</i>	0.02 (+)	<0.0001 (+)	0.05 (–)	0.34 (–)	<0.0001	0.48	Log(Hg + 1) = 0.081 + 0.000139 × Length – 0.001095 × TSI + 0.00225 × wetland area index
<i>Esox masquinongy</i>	0.001 (+)	0.001 (+)	0.59 (–)	0.82 (–)	<0.0001	0.50	Log(Hg + 1) = –0.083 + 0.00256 × wetland area index + 0.00031 × Length

Directional correlations are indicated in parentheses and significance ($\alpha < 0.05$) is indicated in bold

et al. (1994) showed that basin wetlands (which are most similar to the wetlands examined in this study) produced and exported >4 times the MeHg as riverine or valley bottom wetlands. Similarly, Rypel et al. (2008) documented unregulated rivers with large floodplains and connected wetlands had significantly higher Hg concentrations than regulated rivers lacking these landscape features. Thus wetland coverage is a variable that should be used for future field studies examining and predicting Hg bioaccumulation. For bluegill, it is unknown why the rate of change in Hg accumulation with the wetland area index was noticeably slower than for the other species examined. Even though the relationship was significant, it is possible that bioaccumulation effects due to the wetland area are not as pronounced for primary consumer species compared to more predatory species (i.e., the slopes of this relationship may shift upwards through foodwebs). However, this possibility requires future investigation.

Second, strong evidence was found for growth biodilution. Pickhardt et al. (2002) experimentally showed that algal blooms reduced mercury accumulation at higher trophic levels through dilution of mercury in consumed algal cells. Chen and Folt (2005) extended this concept to field data by showing that for northeastern US lakes, Hg concentrations in zooplankton were negatively correlated to phytoplankton densities. For three species in this study, lakes with lower TSI values (more oligotrophic) had higher fish Hg concentrations while lakes with higher TSI values (more eutrophic) had lower Hg concentrations. The two species that did not show a significant biodilution effect were muskellunge, which had a low sample size, and bluegill, which also had a small sample size, a relatively small *P*-value, and again, as a primary consumer may behave differently in terms of Hg bioaccumulation. All species had directional effects strongly suggestive of growth biodilution (i.e., negative correlations). This study therefore provides an additional line of support for growth biodilution in diverse lake types and species.

Mercury concentrations of fishes were not related to ecosystem size for any species, but mean Hg concentrations did scale significantly and positively with ecosystem size across species, i.e., trophic position ($r^2 = 0.89$). Therefore, the assumption that Hg concentrations correlate positively with ecosystem size is accurate, but reflects a pattern mediated by trophic interactions (e.g., Post et al. 2000) rather than by ecosystem size directly. For example, muskellunge (the apex predator in any system it inhabits) had the highest average Hg concentrations of any species examined (all but four populations with small individuals sampled exceeded consumption standards) and also the largest mean ecosystem size. However, this species had high Hg concentrations in small lakes as well as large lakes. Therefore, this and other highly predacious species (e.g., walleye, 75% of lakes

exceeded 0.3 mg kg^{-1}) are likely to have elevated Hg levels in any ecosystem they occupy; however, they more commonly occupy large ecosystems.

There are, therefore, several additional ways to improve fish tissue monitoring. First, current US EPA protocol (USEPA 2000) recommends US states sample one “predator” species per waterbody (in addition to one “bottom-feeding” species). Consumption advisories are then typically based on the results from these two types of fishes. However, the decision over what predator species to sample is clearly a significant one as Hg concentrations of different piscivores can differ by an order of magnitude (Fig. 1). For example, if largemouth bass were sampled alone rather than muskellunge in any of the six lakes where data co-occurred in this study, concentrations would have registered below the recommended consumption limit for four of these six lakes even though Hg concentrations for muskellunge exceeded the 0.3 mg kg^{-1} standard by an average of 3.6 times this benchmark. Tissue monitoring should target species that feed at trophic levels ≥ 4 (e.g., muskellunge, walleye) in ecosystems of any size or type. This is not the standard currently used for many US state tissue monitoring programs. For example, it is difficult to find Hg data for flathead catfish, *Pylodictis olivaris* (another fourth trophic level fish species) even though it is regularly targeted and consumed by commercial, sport, and subsistence fisherman. Furthermore, muskellunge are not sampled for Hg tissue concentrations in every ecosystem in which they commonly occur, but these data suggest that they probably should be. Second, Hg concentrations of fish correlated strongly with fish size, a pattern repeatably observed in most ecosystems (Rypel et al. 2008). If tissue samples for a waterbody are composed of smaller (or even medium-sized) individuals that register near consumption limits, dietary restrictions should be explored for larger fish (if larger fish are known to exist) based on either extrapolations from Hg-to-length regressions of the available data or additional collections of larger individuals. In addition, should models such as the ones presented in this article be used to explore potential Hg levels of fishes in untested lakes, users may wish to experiment with varying fish lengths. Thus, if predicted Hg values of small fish register under consumption limits, but large fish register over consumption limits, such a lake should be tested to determine true Hg levels, especially if angler habits are known (e.g., through a creel survey of similar lakes).

Ecosystem and watershed variation, in addition to, fish size and trophic position affects Hg accumulation by aquatic organisms. Lakes with large wetlands, low productivity, high trophic levels, and large fish typically have elevated fish tissue Hg concentrations. By identifying variation in Hg due to these many factors, one can more accurately estimate Hg concentrations of fish across large landscapes with many

untested waterbodies or anticipate changes in Hg through time due to anthropogenic influences or environmental change (i.e., Vitousek et al. 1997; Lindberg et al. 2007). While these results apply primarily to northern USA and southern Canadian lakes, similar models are likely achievable in other parts of the world with abundant lake ecosystems (e.g., northern Europe, northern Asian and Russian Siberia, central-east Africa). Additionally, there is potential for parallel models to be developed for riverine systems in landscapes where these types of ecosystems are abundant (e.g., central-west Africa, southeast Asia, central America, southern USA). Efforts such as these will be vital to protecting human health as Hg contamination becomes more problematic and more widely recognized as a leading environmental threat to humans across the globe.

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