



Temporal fluctuations in young-of-the-year yellow perch mercury bioaccumulation in lakes of northeastern Minnesota

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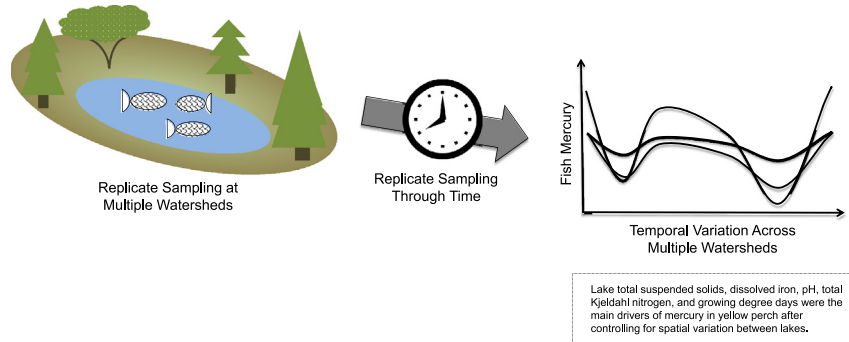
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HIGHLIGHTS

- Investigated temporal variation in young-of-year (YOY) yellow perch mercury concentrations in six lakes over eight years.
- Fish mercury was positively correlated with total suspended solids, dissolved Fe and pH.
- Fish mercury was negatively correlated with total Kjeldahl nitrogen and growing degree days.
- Annual variation in watershed inputs, in-lake processes, and climate can explain temporal patterns in fish Hg.
- Growth biodilution is an important process controlling Hg concentrations.

GRAPHICAL ABSTRACT

Lake total suspended solids, dissolved iron, pH, total Kjeldahl nitrogen, and growing degree days were the main drivers of mercury in yellow perch after controlling for spatial variation between lakes.



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ABSTRACT

Identifying what determines fish mercury (Hg) bioaccumulation remains a key scientific challenge. While there has been substantial research on spatial variation in fish Hg bioaccumulation, the factors that influence temporal fluctuations in fish Hg have received less attention to date. In this study, we built upon a growing body of research investigating young-of-the-year (YOY) yellow perch Hg bioaccumulation and investigated annual fluctuations in YOY yellow perch Hg in six lakes in northeastern Minnesota over eight years. After accounting for spatial variation between the study lakes, we used model averaging to identify the lake physiochemical and climate factors that best explain temporal variation in fish biomass and fish Hg. Fish biomass of YOY yellow perch had a positive relationship with chlorophyll- α and total Kjeldahl nitrogen and a negative relationship with dissolved iron and dissolved oxygen. There was a positive relationship between annual variation in yellow perch Hg concentration and annual variation in lake total suspended solids, dissolved Fe and pH. Additionally, there was a negative relationship between fish Hg concentration and lake total Kjeldahl nitrogen and growing degree days. Together, our results suggest that annual variation in allochthonous inputs from the watershed, in-lake processes, and climate variables can explain temporal patterns in Hg bioaccumulation and growth biodilution is an important process controlling yellow perch Hg concentrations.

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1. Introduction

Consumption of fish with elevated mercury (Hg) concentrations poses a health risk to both humans and wildlife. Identifying where and when fish Hg concentrations exceed limits established for human and wildlife health is a key challenge for policy makers and land managers. While the spatial controls on fish Hg bioaccumulation have been well studied (e.g., Cope et al., 1990; Gabriel et al., 2009; Yu et al., 2011), the controls over seasonal and annual fluctuations within lakes have received far less attention.

The bioaccumulation of Hg in fish reflects the influence of watershed sources of Hg to surface waters, chemical and biological controls over Hg methylation rates in lakes and adjacent wetlands, as well as fish diet (Greenfield et al., 2001; Dittman and Driscoll, 2009). Multi-lake studies show strong spatial correlations between elevated Hg concentrations in low trophic-status fish and upland Hg stocks (Gabriel et al., 2009), as well as watershed and lake characteristics that impact the abundance and production of methyl Hg by microbes (e.g., connected wetlands and lake pH) (Wiener et al., 2006). In addition to factors that influence the availability and abundance of Hg for bioaccumulation, fish diet and growth “biodilution” of tissue Hg also influence fish Hg concentrations (Karimi et al., 2007). For example, researchers have observed negative relationships between lake average fish Hg concentrations and available nutrients (i.e., total P) when comparing across lakes: more eutrophic or productive lakes lead to greater fish mass and lower tissue Hg concentrations per unit mass (Swain and Helwig, 1989; Chen and Folt, 2005; Karimi et al., 2007).

In addition to the spatial variation in fish Hg concentration, fish Hg bioaccumulation fluctuates within and between seasons (Monson and Brezonik, 1998; Monson, 2009). Temporal fluctuations in both natural and anthropogenic processes, such as climate, atmospheric deposition, and upland runoff, can impact temporal variability in fish Hg bioaccumulation. For example, changes in lake water level (Sorensen et al., 2005; Willacker et al., 2016; Riggs et al., 2017), algal biomass in response to eutrophication (Chen et al., 2012), and atmospheric Hg deposition (Brigham et al., 2014) have all been identified as potential drivers of temporal variation in Hg bioaccumulation in fish and other lake biota. Unfortunately, the multi-year, continuous studies of fish Hg and associated lake chemistry data necessary to understand these temporal changes are rare. There

have been few quantitative analyses of interannual variation in fish Hg bioaccumulation or the factors driving such variation.

We addressed this gap in knowledge by evaluating Hg concentration and biomass in young-of-the-year (YOY) yellow perch (*Perca flavescens*) continuously for eight years in six small, shallow lakes in the Boundary Waters Canoe Area Wilderness (Superior National Forest, Minnesota). Previously, these six lakes contributed to a short-term (two year) study of the spatial controls over yellow perch Hg among a larger group of lakes in the Boundary Waters Canoe Area Wilderness (Gabriel et al., 2009). This paper presents, for the first time, the data from years three through eight. Additionally, this study examines temporal annual variability, as opposed to spatial variability among lakes, in fish Hg.

Young-of-the-year yellow perch are an excellent organism to monitor for temporal change in Hg bioaccumulation. Young yellow perch feed primarily on zooplankton during their first year (Whiteside et al., 1985) and are indicators of aquatic food web Hg status; Hg concentration in YOY yellow perch is a strong predictor of Hg concentration in co-located piscivorous fish and wildlife (Cope et al., 1990). Additionally, yellow perch Hg concentration is strongly influenced by allochthonous inputs of organic matter and Hg from the surrounding watershed in our study lakes. Previously, Gabriel et al. (2009) found that total Hg in the upland forest floor, watershed area, and lake iron concentration were the strongest spatial predictors of YOY yellow perch total Hg. Consequently, we hypothesized that inter-annual variation in YOY yellow perch Hg bioaccumulation would be driven by temporal fluctuations in allochthonous inputs from the watershed, in-lake processes, and climate, and expect an opposite relationship between fish biomass and fish Hg concentration (i.e. growth biodilution).

2. Material and methods

2.1. Site description

We investigated YOY yellow perch Hg and lake chemistry in six small lakes in the Boundary Waters Canoe Area Wilderness (Superior National Forest) in northeastern Minnesota (Fig. 1). Lake chemistry, morphometry, and watershed characteristics differ among the lakes (Supplement, Table S1). The lakes, which range from 5.2 to 15.2 m deep and 16.5 to 153.3 ha in area, are headwater lakes that drain

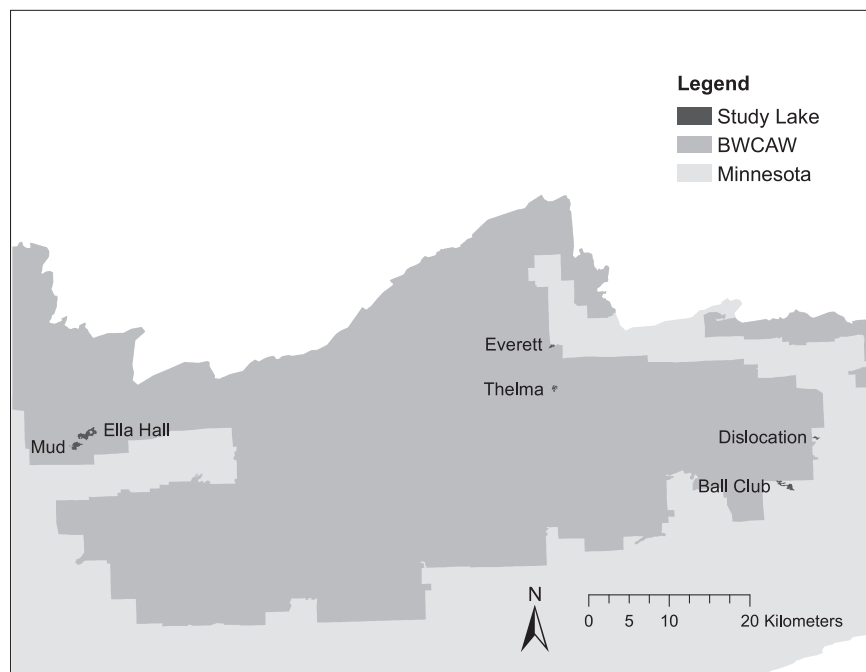


Fig. 1. Map of study lake locations in the Boundary Waters Canoe Area Wilderness (BWCAW), Superior National Forest, northeastern Minnesota.

surrounding watersheds 64.2 to 621.4 ha in area. The upland soils in the lake watersheds are relatively shallow Inceptisols over bedrock (Roger Risley, USDA Natural Resources Conservation Service, *personal communication*). Across the lakes during the study period (2004–2012), mean annual temperature was 3.2 °C and mean annual precipitation was 728 mm yr⁻¹, with approximately 66% of the precipitation falling during spring and summer (April–September).

2.2. Fish sampling and calculation of biomass-adjusted fish Hg concentration

Yellow perch that hatched the previous spring (i.e., age 1 or young-of-the-year) were targeted during collection. We verified fish age using observed fish size (length vs. mass) relationships within each lake and year, as well as scale data (Schneider et al., 2000; Schneider, 2001). Yellow perch were sampled annually in 2005 through 2012 via electroshocking once the study lakes were accessible following ice-out (Supplement, Table S2). Immediately after collection, fish total length and mass were measured and fish scales were subsampled. Whole fish samples were transported to the lab on ice where they were digested in concentrated nitric acid at 70 °C overnight in PTFE bombs. An aliquot of the digest was analyzed for tissue total Hg using the double gold amalgamation method of Bloom and Creclius (1983) (similar to US EPA method 1631) with a Brooks Rand Model III cold vapor atomic fluorescence spectroscopy (CVAFS) analyzer and Mercury Guru software. At the start of each analysis, standard curves were run (mean R²: 0.9994). The average difference between analytical duplicates (relative percent difference) was 4.17%; average percent recovery of analytical standards, NIST reference material 2976 (freeze-dried mussel tissue), and matrix spike recoveries were 101.1%, 101.8%, and 97.6%, respectively. The limit of detection for the analysis was 0.045 ng.

2.3. Water sampling

Lake water samples were collected from each study lake multiple times per year during the frost-free periods (approximately June–September) of 2004 through 2012. We used a Van Dorn sampler to collect lake water samples at 1 m depth at an index station over the deepest spot of the lake's main basin. Additional in-lake parameters (specifically, lake water pH, dissolved oxygen concentration, and temperature) were sampled using a water quality sonde at 1 m depth. Also, Secchi depth and lake water level were recorded (the latter in relation to a fixed reference object). Non-acidified water samples were analyzed for nutrients (total phosphorus and total Kjeldahl nitrogen), total suspended solids, and other organics (total carbon and chlorophyll- α ; methods: EPA 365.3, EPA 351.2, USGS I-3765-85, SM 5310C, and SM 10200, respectively). Additionally, non-acidified samples were analyzed for dissolved iron (Fe) and sulfur (S) via inductively coupled plasma optical emission spectrometry.

In 2008 through 2012, an additional unfiltered lake water sample was collected for total Hg analysis. The samples were preserved in the field with bromine monochloride (BrCl; a strong oxidizer) and digested (heated to 70 °C overnight) prior to analysis. The digestate was added to a bubbler flask and excess BrCl was reduced with hydroxylammonium hydrochloride. Total Hg analysis was performed as described above using CVAFS. The limit of detection was 0.049 ng (mean plus three times the standard deviation of the analytical blanks); average standard curve fit was 0.9994 (R²); average relative percent difference between analytical duplicates was 6.5%; and average recovery of analytical standards, NIST reference material 1515 (apple leaves), and matrix spikes were 98.9%, 99.8%, and 96.9%, respectively.

2.4. Climate data

Monthly precipitation totals at each lake were calculated from the Minnesota State Climatology Gridded Precipitation database, which are interpolated estimates from local precipitation gauge records

(Minnesota State Climatology Office, 2015). Minimum and maximum daily temperature records are from the PRISM climate model (PRISM Climate Group, 2004). We calculated seasonal metrics of total precipitation and average temperature in winter (January–March), spring (April–June), summer (July–September), and fall (October–December) for 2003–2012. We also calculated four annual temperature metrics that reflect the influence of climate on fish productivity (Chezik et al., 2013): growing degrees (sum of growing season degrees > 10 °C), growing degree days (total number of growing season days during which mean daily temperature > 10 °C), freezing degrees (sum degrees < 0 °C between growing seasons), and freezing degree days (number of days between growing seasons when mean daily temperature < 0 °C).

2.5. Statistical analyses

All analyses were performed in R (R version 3.3.1) and significance was evaluated at $\alpha = 0.05$. First, we tested for temporal trends in fish mass (g) and fish Hg concentration (ng g⁻¹) utilizing the Mann-Kendall trend test (*Kendall* package in R). We evaluated the interactive effects of year (continuous variable) and lake identity (discrete variable) on fish mass and Hg concentration using two-way ANOVA (Type II ANOVA; *car* package). The significant interaction between year and lake was evaluated with post-hoc comparisons (*lsmeans* package); *p*-values were Bonferroni corrected for multiple comparisons.

Next we evaluated the effects of climate and lake physiochemical parameters on YOY yellow perch mass and Hg concentration using a mixed-effects statistical model (*lme4* package) and model averaging (Grueber et al., 2011). Model averaging is a model selection approach that identifies the set of equivalent sub-models that best fit the data from the largest possible model that includes all possible predictors. We designated sub-models as equivalent if the difference in Akaike Information Criterion (AIC; a measure of model fit) among them was <4 (*dredge* function in the *MuMIn* package). Once we selected the set of best, equivalent sub-models, we evaluated the importance and significance of the predictor variables included in this top set of sub-models. Reported *p*-values are from the full model-averaged coefficients (e.g., across all possible models, including the ones where the parameter was absent), which is a better estimate of which factors have the strongest effects on the response variable (Grueber et al., 2011).

Model averaging selected equivalent sub-models from the largest possible model that included all possible predictors. The predictor parameters evaluated included mean annual lake physiochemical variables and summer climate variables from the previous year (the year the fish hatched), as well as winter climate variables from the first winter of growth. We tested for multicollinearity among these predictors by calculating the variance inflation factor (VIF) of each predictor in the largest possible model. A single variable was chosen when multicollinearity was encountered. Additionally, we did not include lake water Hg as a predictor due to the low number of years sampled ($n = 4$) compared to the other parameters. Variables were ln-transformed as needed to meet model assumptions of normality. Site crossed with year was included as a random effect in all models. In the final “averaged” model (i.e., the set of sub-models that fit the data best and were equivalent), the variance explained by the fixed effects only (marginal R²) and the variance explained by both fixed and random effects (conditional R²) was calculated using the *r.squaredGLMM* function (*MuMIn* package; Nakagawa and Schielzeth, 2013).

A constraint of the model structure used in the model averaging analysis is that it may not reveal the effects of predictors on temporal variation in fish weight or Hg when the bivariate relationship is opposite when examining within lakes (i.e., temporal variation) versus across lakes (i.e., spatial variation). To examine these temporal predictors of fish Hg and fish weight, we also analyzed the effects of temporal variation in climate and lake physiochemical variables on YOY yellow perch mass and Hg concentration within each study lake. Specifically, we evaluated the bivariate relationship between these predictor

parameters and either fish mass or Hg concentration separately for each study lake. We calculated Pearson correlation coefficients (*Pearson DS* package) to visualize these bivariate relationships and used mixed-effects statistical models (*nlme* package) to test their significance. In the statistical model, year was included in the model as a random effect and *p*-values for Bonferroni corrected for multiple comparisons. Finally, we tested the significance of the relationship between fish weight and fish Hg within each lake using regression analysis. To approximate normal distributions variables were ln-transformed as needed to meet model assumptions of normality.

3. Results

There were no significant temporal trends in YOY yellow perch mass and Hg concentration from 2005 to 2012 in any of the study lakes ($p > 0.1$ for all; Supplement, Table S3). Despite the lack of temporal trends, there was a significant interactive effect of lake identity and year on both fish mass and Hg concentration in our study lakes ($p < 0.0001$, $R^2 = 0.80$ and $p < 0.0001$, $R^2 = 0.83$, respectively; Figs. 2 and 3).

Model averaging identified the lake physiochemical and climate variables that best explained the overall variation in fish mass and Hg concentration in our study lakes. Lake water chlorophyll- α , total Kjeldahl nitrogen, dissolved Fe, and dissolved oxygen concentrations were the strongest predictors of fish mass (Table 1; $p < 0.001$ for all). Fish mass increased with increasing concentrations of chlorophyll- α and total Kjeldahl nitrogen and decreasing concentrations of Fe and dissolved oxygen. The strongest predictors of fish Hg concentration were lake water total suspended solids, lake water Fe, lake water pH, lake water total Kjeldahl nitrogen, and growing degree days (Table 2; $p < 0.01$ for all). Fish Hg concentration increased as the concentrations of lake water Fe and total suspended solids increased, as lake pH increased, as lake

total Kjeldahl nitrogen decreased, and the number of growing degree days decreased.

There were few consistent relationships between either fish mass or fish Hg concentration and lake physiochemical and climate variables within each of the study lakes (data not shown). Additionally, only one of these relationships was statistically significant: there was a significant positive relationship between YOY yellow perch Hg concentration and lake water Hg concentration during the previous growing season in Dislocation Lake ($p < 0.01$). Finally, we observed a significant negative exponential relationship between fish Hg concentration and fish mass at four of the six study lakes: as the mass of the sampled fish increased, Hg concentration decreased exponentially at Mud, Thelma, Ball Club, and Dislocation Lakes (Fig. 4).

4. Discussion

4.1. Lake productivity, dissolved oxygen, and dissolved Fe influenced variation in young-of-the-year yellow perch biomass

The strongest predictor of YOY yellow perch biomass across our small, shallow study lakes in northeastern Minnesota was lake chlorophyll- α . The relationship between lake primary production and fish productivity is well established (Downing et al., 1990; McInerney and Cross, 1999); this relationship reflects the influence of nutrient availability on the productivity and growth of this low trophic level fish species through algal and/or zooplankton biomass. Similarly, total Kjeldahl nitrogen is indicative of lake productivity with productivity increasing with increasing concentrations of nitrogen (Elser et al., 2007). We also found a negative relationship between yellow perch mass and dissolved oxygen which has been shown in other studies (Carlson et al., 1980). At low dissolved oxygen levels fish decrease their metabolic rate to compensate thereby reducing their growth. Although we

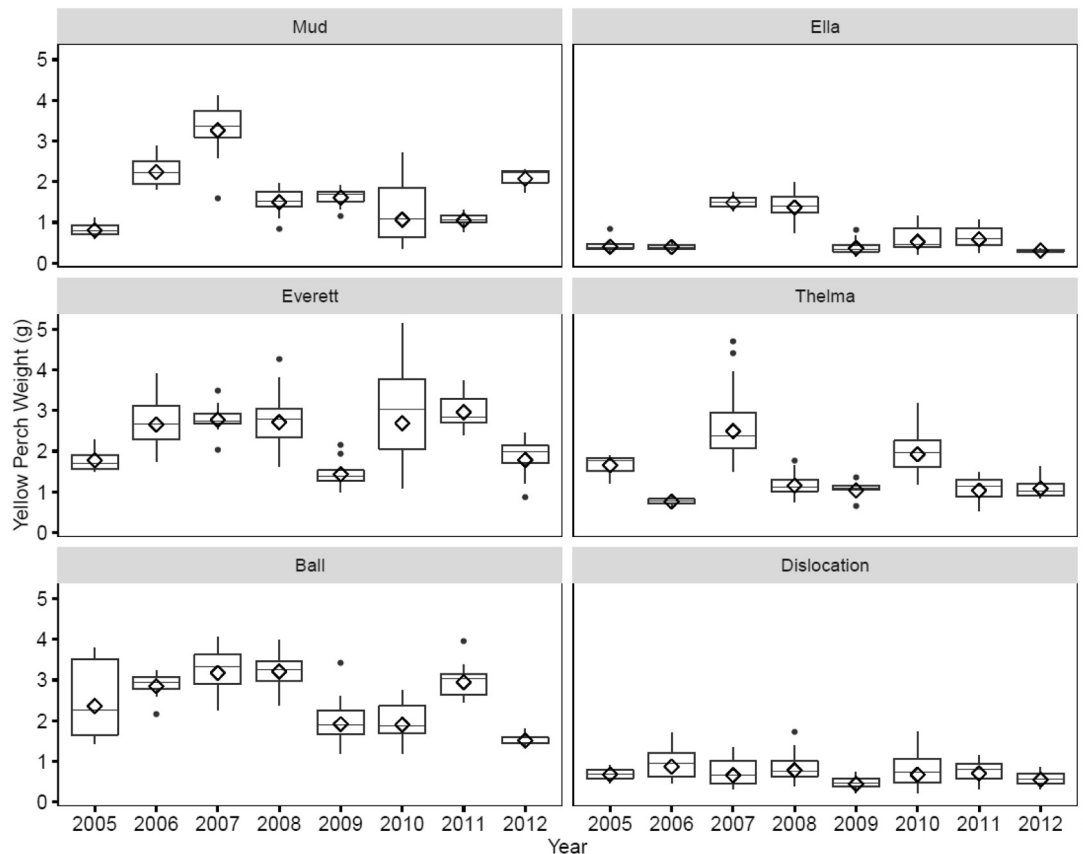


Fig. 2. Annual variation in young-of-the-year yellow perch fish mass (g) in six study lakes. All panels show geometric mean (diamond symbol) and ranges of fish sampled in 2005–2012 at all lakes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

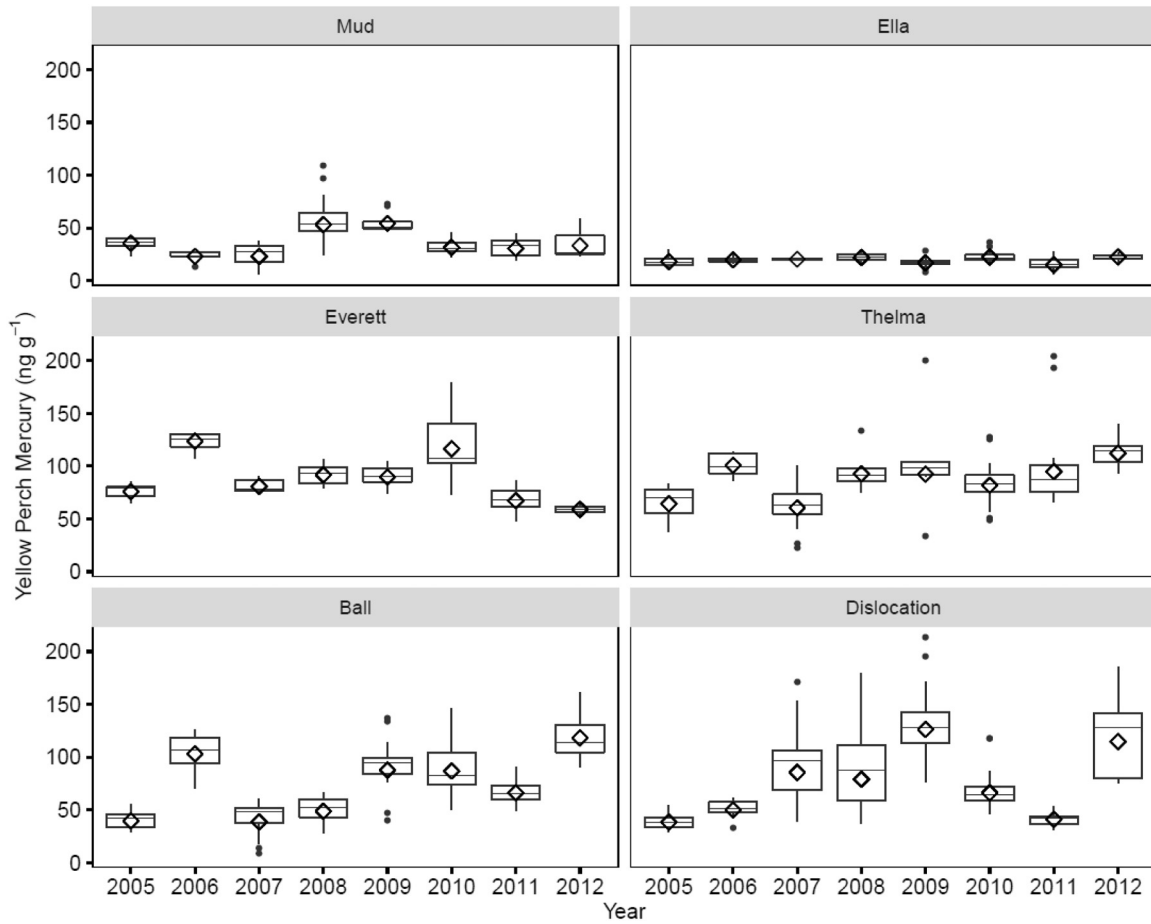


Fig. 3. Annual variation in young-of-the-year yellow perch fish mercury (Hg) concentration (ng g^{-1}) in six study lakes. All panels show geometric mean (diamond symbol) and ranges of fish sampled in 2005–2012 at all lakes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

didn't find any studies that specifically assessed how dissolved Fe concentrations influence perch growth, laboratory studies indicate high dissolved Fe concentrations can have negative impacts on grayling (*Thymallus thymallus*) survival (Vuorinen et al., 1998). Although we anticipated that within lake analyses would lead to significant relationships between climate and lake chemistry parameters and yellow perch biomass, this was not supported by our results.

4.2. Climate, in-lake, and watershed influences on variability in yellow perch Hg bioaccumulation

Variation in lake total suspended solids and lake dissolved Fe were the strongest predictors of fish Hg concentration: as lake total suspended solids and Fe increased, fish Hg concentration increased. The influence of lake total suspended solids on fish Hg concentration suggest that annual variability in allochthonous inputs of organic matter

Table 1
Parameters from averaged models predicting fish mass. Table shows the average β across all models, p -value and the number of models containing the parameter.

Model parameter	Average β coefficient	p -Value	Number of models containing parameter
Lake chlorophyll- α †	0.097	<0.05	89
Lake dissolved oxygen†	-0.236	<0.05	89
Lake dissolved iron†	-2.99	<0.05	89
Log Lake total Kjeldahl nitrogen†	0.367	<0.05	89
Lake total phosphorus†	-0.148	0.21	64
Lake water total sulfate†	-0.074	0.32	54
Freezing degrees	-0.0005	0.37	58
Freezing degree days	0.006	0.46	48
Lake pH†	0.032	0.62	37
Lake total suspended solids†	-0.030	0.65	32
Growing degrees ‡	0.0002	0.80	28
Growing degree days ‡	-0.001	0.84	24

† Mean annual lake variables from fish first growing season (one year prior to fish sampling year).

‡ Growing degrees and growing degree days from fish first growing season (one year prior to fish sampling year).

Table 2
Parameters from averaged models predicting fish Hg concentration. Table shows the average β across all models, p -value and the number of models containing the parameter.

Model parameter	Average β coefficient	p -Value	Number of models containing parameter
Growing degree days ‡	-0.034	<0.05	42
Lake dissolved iron†	4.554	<0.05	42
Lake pH†	0.170	<0.05	42
Log Lake total Kjeldahl nitrogen†	-0.439	<0.05	42
Lake total suspended solids†	0.704	<0.05	42
Lake dissolved oxygen†	0.101	0.07	36
Growing degrees‡	-0.001	0.37	25
Lake total phosphorus†	2.874	0.63	16
Lake chlorophyll- α †	-0.003	0.70	16
Lake water total sulfate†	-0.009	0.75	12
Freezing degree days	0.001	0.78	13
Freezing degrees	0.00006	0.82	12

† Mean annual lake variables from fish first growing season (one year prior to fish sampling year).

‡ Growing degrees and growing degree days from fish first growing season (one year prior to fish sampling year).

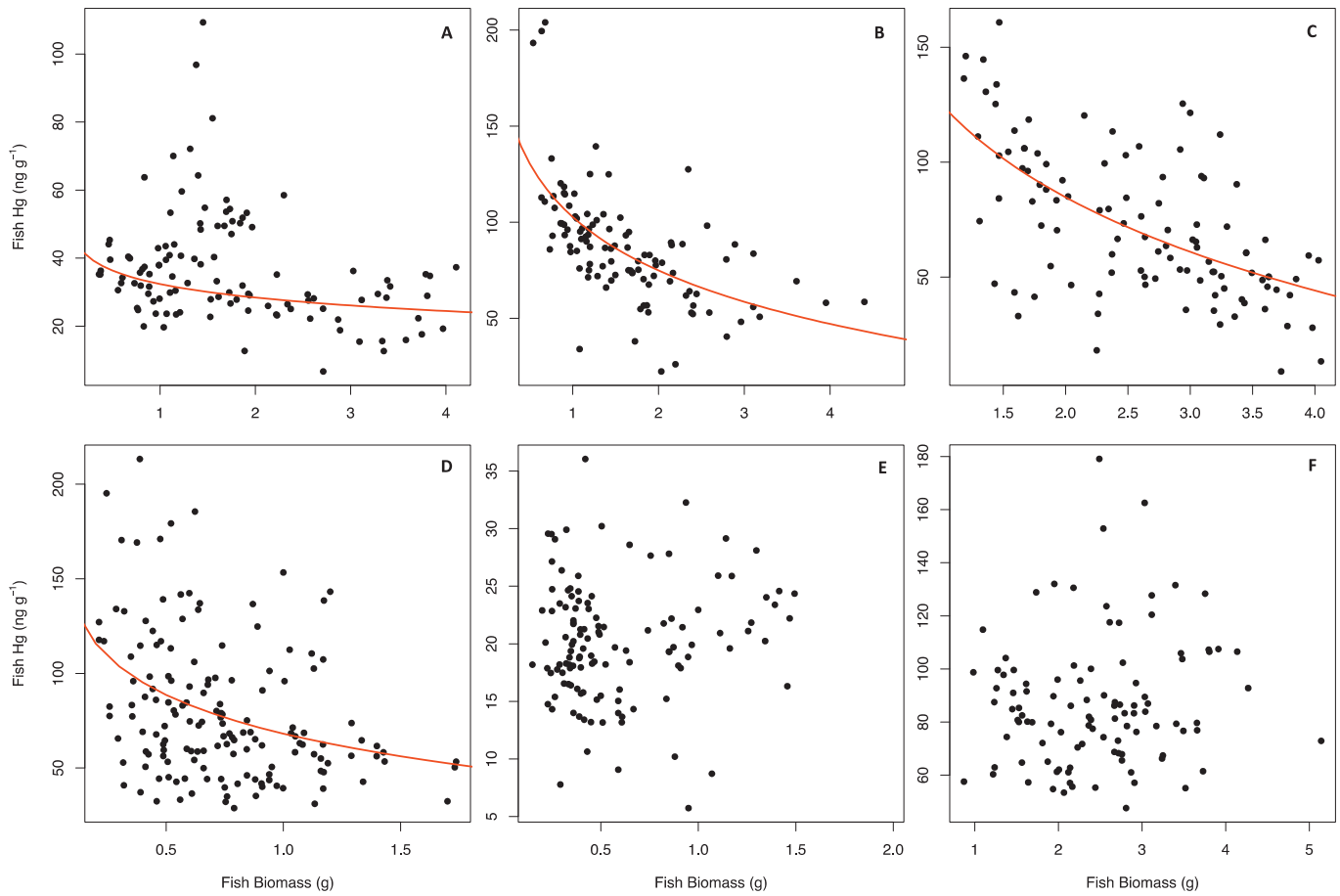


Fig. 4. Relationship between young-of-the-year yellow perch Hg concentration and mass in Mud (a), Thelma (b), Ball Club (c), Dislocation (d), Ella Hall (e), and Everett (f) lakes. There was a significant negative exponential relationship between fish Hg concentration and mass in Mud, Thelma, Ball Club, and Dislocation lakes; there was no statistically significant relationship between fish Hg concentration and mass in Ella Hall and Everett lakes. The significant negative exponential relationships were: Mud: $y \sim (-5.73 * \log(\text{Biomass})) + 32.39$, $p < 0.0001$, $R^2 = 0.19$; Thelma: $y \sim (-40.14 * \log(\text{Biomass})) + 102.71$, $p < 0.0001$, $R^2 = 0.38$; Ball Club: $y \sim (-58.53 * \log(\text{Biomass})) + 125.27$, $p < 0.0001$, $R^2 = 0.37$; Dislocation: $y \sim (-29.52 * \log(\text{Biomass})) + 68.17$, $p < 0.0001$, $R^2 = 0.13$. To compute mass-adjusted fish Hg concentration, we divided the unadjusted fish Hg concentration by the linear regression equation (i.e., $mx + b$) when significant; Ella Hall and Everett lake fish Hg concentration was divided by the overall lake mean (20.13 and 86.88 ng g^{-1} , respectively). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and Hg to the lakes from the surrounding watershed drives temporal variation in fish Hg concentration. However, positive relationships with dissolved Fe may indicate in-lake processes also have an important role. Although the source of Fe is likely from weathering of soil minerals in the watershed, resuspension from lake sediments and subsequent iron reduction by microbes can lead to higher methyl mercury production in the water column (Chadwick et al., 2006; Fleming et al., 2006). The positive relationship with lake pH and fish Hg concentration is surprising considering many studies have shown negative relationships with lake pH and fish Hg concentration (e.g. Grieb et al., 1990; Driscoll et al., 1994; Wiener et al., 2006). However, those studies have mainly assessed low pH acid sensitive lakes with few studies occurring in near neutral pH lake systems such as this study (Table S1). A recent study across a gradient of north temperate to arctic lakes that span a pH gradient from 6.6–8.3 found no significant relationship with lake pH and mercury in invertebrates and even found a non-significant positive trend for pH and *Chironomids* methyl mercury concentration (Chetelat et al., 2018). From what little literature exists for higher pH environments, it appears that the negative pH-biota Hg concentration relationship is not supported, perhaps because of higher binding capacity of dissolved organic carbon and Hg at higher pH and subsequent lower methylation rates (Miskimmin et al., 1992; Ravichandran, 2004). In line with our results, Lange et al. (1993) found a negative correlation between total N and mercury in largemouth bass (*Micropterus salmoides*) in Florida. They attributed the negative correlation to more

eutrophic lakes leading to greater fish growth rates and subsequent growth biodilution. Similarly, greater growing degree days leads to greater fish growth rates and growth biodilution of Hg (Rennie et al., 2010). The negative relationship between fish Hg concentration and fish mass at four of the study lakes suggest that at these lakes factors that increase fish growth (such as lake nutrients, discussed above) led to a decrease in the concentration of Hg in fish tissue. A number of studies have observed an impact of growth biodilution on fish Hg concentration. For example, nutrient concentration across lakes often predicts fish Hg concentration. In a study of Minnesota lakes, Heiskary and Helwig (1986) found that fish Hg concentration was negatively correlated with lake total P concentration, among other lake variables. Additionally, Essington and Houser (2003) found that Hg concentration in YOY yellow perch was 30–40% less in nutrient enriched lakes relative to control lakes in a whole lake nutrient addition study.

Interestingly, growth biodilution was apparent in some, but not all of our study lakes (Fig. 4). The presence or absence of the relationship is not readily explained by differences in lake size or other lakes characteristics. For example, in the two smallest lakes by area, growth biodilution was apparent in one lake (Dislocation) but not the other (Everett). Although we do not know the causation for growth biodilution, we suspect that differences in lake trophic structure and fish diet may drive this pattern. For example, yellow perch are known to feed on a variety of zooplankton food sources during their first year (Whiteside et al., 1985); zooplankton community composition could vary between the lakes. Alternately, maternal

effects on fish Hg concentration at hatching (i.e., Hg content in fish eggs) could influence the concentration of Hg in newly hatched yellow perch, again leading to lake by lake differences that subsequently influence the biodilution of Hg as the fish grows during its first year.

4.3. Conclusion

Our results support our prediction of a relationship between annual variation in YOY yellow perch Hg and, watershed influences (TSS), lake chemistry (dissolved Fe, pH and total Kjeldahl nitrogen), and climate (growing degree days). Specifically, temporal fluctuations in lake total suspended solids may reflect variation in allochthonous inputs to lakes while dissolved Fe, pH and total Kjeldahl nitrogen are more reflective of in-lake processing. The negative relationship between fish Hg concentration and lake total Kjeldahl nitrogen and growing degree days suggests that lake nutrient status and longer growing conditions leads to biodilution of Hg in fish. It is well known that increased lake productivity can decrease the fish Hg concentration via growth biodilution (Karimi et al., 2007). Consequently, in addition to the effect of annual changes in watershed inputs to these small shallow lakes, air temperature, lake productivity, and lake nutrients also influence temporal fluctuations in fish Hg bioaccumulation via their influence on fish mass.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.11.280>.

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