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Climate and Physiography Predict Mercury Concentrations in Game Fish Species in Quebec Lakes Better than Anthropogenic Disturbances

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Abstract The fluctuations of mercury levels (Hg) in fish consumed by sport fishers in North-Eastern America depend upon a plethora of interrelated biological and abiological factors. To identify the dominant factors ultimately controlling fish Hg concentrations, we compiled mercury levels (Hg) during the 1976–2010 period in 90 large natural lakes in Quebec (Canada) for two major game species: northern pike (*Esox lucius*) and walleye (*Sander vitreus*). Our statistical analysis included 28 geographic information system variables and 15 climatic variables, including sulfate deposition. Higher winter temperatures explained 36 % of the variability in higher walleye growth rates, in turn accounting for 54 % of the variability in lower Hg concentrations. For northern pike, the dominance of a flat topography in the watershed explained 31 % of the variability in lower Hg concentrations. Higher mean annual temperatures explained 27 % of the variability in higher pike Hg concentrations. Pelagic versus littoral preferred habitats for walleye and pike respectively could explain the contrasted effect of temperature between the two species. Heavy logging could only explain 2 % of the increase in walleye Hg concentrations. The influence of mining on fish Hg concentrations appeared to be masked by climatic effects.

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Mercury (Hg) bioaccumulation in wild, freshwater fish potentially consumed by human populations remains a subject of deep concern for vulnerable populations and public health authorities. In Canada, the United States, and Feno-Scandinavia particularly, fish consumption advisories are mostly related to high levels of Hg in fish flesh (Health-Canada; UNEP 2013; US-EPA). There are a plethora of studies attempting to explain the environmental conditions leading to higher Hg concentrations in fish. A first group of studies evaluates the importance of a series of natural, environmental factors that influence Hg concentrations in fish. Many items on this long list of factors are related to the nature of the watershed, including the abundance of wetlands (Burns et al. 2012; Greenfield et al. 2001; St. Louis et al. 1994, 1996), mean watershed slopes (Moingt et al. 2013; Teisserenc et al. 2011), presence of beaver dams in the watershed (Roy et al. 2009), drainage area to lake area ratios (Roué-LeGall et al. 2005), and vegetation types in the watershed (Drenner et al. 2013; Graydon et al. 2012; Moingt et al. 2013). Others factors include physicochemical properties of the aquatic systems, such as pH (Greenfield et al. 2001) and dissolved organic carbon (DOC) (Burns et al. 2012; Golden et al. 2012), as well as biological conditions of the fish populations such as the structure of the food chain (Gantner et al. 2010b; Henderson et al. 2004), and fish growth rates (Greenfield et al. 2001; Lavigne et al. 2010; Simoneau et al. 2005).

More recently, additional complexity has been added by a second group of Hg studies of anthropogenic activities, which potentially play significant roles in long-term dynamics of Hg bioaccumulation in fish. In this vein, there are studies of anthropogenic Hg atmospheric deposition (Harris et al. 2007) and land-use and land-use changes including logging (Garcia and Carignan 2000; Porvari et al. 2003; Sørensen et al. 2009), reservoir impoundment (Bodaly et al. 2007; Lucotte et al.

1999a), wetland drainage (Holmes and Lean 2006), and mining (Weech et al. 2004; Winch et al. 2009). There also are studies of alterations in food web dynamics by species addition (Johnston et al. 2003), removal (Lepak et al. 2009), or through intensive fishing (Surette et al. 2006), as well as research on eutrophication through agriculture (Sonesten 2003), urbanization (Eckley et al. 2008), or aquaculture (Meng et al. 2010). In addition, it is now suspected that lake acidification (Hrabik and Watras 2002; Lacoul et al. 2011; Rennie et al. 2005; Scheuhammer and Graham 1999) and climate change (Rennie et al. 2010; Verta et al. 2010) may influence to some extent Hg concentrations in fish.

All natural and anthropogenic factors known to influence Hg concentrations in fish interact with one another, thus making the pattern of Hg accumulation in fish very confusing in relation to any one dominant factor. The absence of regular and coherent monitoring of Hg dynamics in large lakes at a regional scale and over several decades adds to the difficulty of establishing reliable temporal and spatial trends for fish Hg concentrations. Indeed, fish Hg concentrations may be drastically different among nearby lakes located in the same eco-region even when sampled during a single field campaign (Lucotte et al. 1995) or may greatly fluctuate from year to year in a given lake (Johnston et al. 2003; Monson et al. 2011; Weis 2004). It also is very difficult to definitively pinpoint a particular anthropogenic activity as responsible for enhancing the bioaccumulation of Hg in fish over several years (Monson et al. 2011; Neff et al. 2013; Wyn et al. 2010). These are causes of inconsistencies among attempts to publish sound recommendations for fishers and fish eating humans.

Modelers have attempted to integrate as many factors as possible in Hg preference models (Roué-LeGall et al. 2005), Hg regression models (Depew et al. 2013; Gantner et al. 2010a, b; Greenfield et al. 2001; Monson et al. 2011) or Hg mechanistic models (Golden et al. 2012; Harris et al. 2007). The latter models have mostly been calibrated with data collected in small headwater lakes and thus do not appear to predict fish Hg concentrations in larger lakes with accuracy. Hg dynamics in those large aquatic systems are characterized by a relative inertia in responding to a great number of interacting factors, often difficult to measure at a large geographical scale (Moingt et al. 2013).

In this paper, we mined data related to Hg concentrations over the last three decades in two predatory game fish species, living in natural lakes in Quebec (Canada). We did not consider Hg data in fish living in large man-made reservoirs, Hg dynamics in these environments being radically modified by the impoundment (Lucotte et al. 1999a; Montgomery et al. 2000). We chose to report on Hg concentrations in two fish species among the most sought after by fish eating humans, walleye (*Sander vitreus*), a pelagic species and northern pike (*Esox lucius*), a littoral species. We compiled readily available variables, as well as those that could be deducted from geospatial information, considering variables found to be related to Hg dynamics in previous scientific studies. We opted for multiple linear regression models using stepwise variable selection in order to produce a first synoptic interpretation of the respective influence of natural and anthropogenic factors on fish Hg concentrations. By delineating the environmental conditions leading to the highest Hg concentrations in fish, our findings contribute to improved management of health risks associated with wild freshwater fish consumption in the boreal forest region. In a complementary way, because our models consider land-use changes and climate change over time, they represent a useful tool for public health authorities and fish consumers to better anticipate the potential impacts of sustained anthropogenic activities on the quality of wild freshwater fish.

Materials and Methods

Building a Comprehensive Data Set

The compilation of data repositories of the Quebec Ministry of Environment, Hydro-Quebec, COMERN NSERC strategic network, and Environment Canada's CARA research program allowed us to construct our total Hg concentration database for walleye and northern pike caught during the 1976–2010 period (Table 1). Methods for total Hg analysis in fish are described in Lucotte et al. (1999b) and Moingt et al. (2013). Our study covered a total of 90 natural lakes (not including manmade reservoirs) stretching over a large latitudinal span of the boreal forest domain in mid-northern Quebec, Canada (46.26–53.28°N). Contrary to other published studies on temporal and geographical Hg trends in lakes, most lakes considered in our study are large lakes (mean surface area of 59.3 km^2; range 0.34–951 km^2), with extended watersheds (mean land area draining directly into the lake, without transiting through another lake, of 474 km^2, range 2.1–5974 km^2) and where urban anglers, sports fishers, and members of first nations (i.e., aboriginal peoples of Canada) are accustomed to fishing. All together, we built a matrix including 44 variables (corresponding to 4862 entries) describing fish populations (with Hg concentrations and ages calculated at standard length), physiographical settings, vegetation cover in the drainage basin, and climatic conditions (temperature and precipitation) at the time of fish sampling as well as anthropogenic activities potentially influencing Hg dynamics (mining, logging, and acidification; Tables 2 and 3).

To compare different years and lakes, we calculated fish population total Hg levels at standard lengths using the criteria described in detail by Lavigne et al. (2010).

Table 1 Number of lakes sampled once, twice, and three times for determination of Hg concentrations in walleyes and pikes and for fish ages measurements

	Total number of lakes	Number of lakes sampled twice	Number of lakes sampled three times	Total number of sampling
Hg concentrations in walleyes	73	9	3	88
Age measurements of walleyes	38	7	3	52
Hg concentrations in pikes	71	10	2	85
Age measurements of pikes	24	12	0	36

Table 2 Variables deducted from geospatial images

	Watershed level	Units	Range
Longitude	3rd	°W	79.5–73.8
Latitude (LAT)	3rd	°N	46.3–53.3
Lake order	1st	Number	0–6
Lake area (LA)	–	Km^2	0.34–951
Watershed area (WA)	3rd	Km^2	2.07–5974
WA/LA	3rd	Ratio	1.35–54.3
Drainage length to the lake	3rd	Km	0–2374
Drainage density	3rd	Km^{-1}	0–10.0
Clay substratum (CS)	3rd	% WA	0–100
Mean slope of watershed (MS)	3rd	%	0.81–9.9
0–2 % slope of watershed	3rd	% WA	12.8–84.7
2–6 % slope of watershed	3rd	% WA	9.49–38.6
6–10 % slope of watershed	3rd	% WA	1.82–53.4
10–20 % slope of watershed	3rd	% WA	0.2–23.4
>20 % slope of watershed (SL20)	3rd	% WA	0–13.9
Forest	3rd	% WA	30.4–95.3
Deciduous forest	3rd	% WA	0–20.5
Mixed forest <50 % coniferous (MF)	3rd	% WA	0.62–50.0
Mixed forest >50 % coniferous	3rd	% WA	0.52–41.4
Coniferous, well-drained soils	3rd	% WA	3.17–47.0
Coniferous, poorly drained soils (CPDS)	3rd	% WA	0.75–69.5
Wetland	3rd	% WA	0–49.1
Wetland (WB)	4th	% WA	0–46.0
Bare soils (BS)	3rd	% WA	0–50.7
Herbaceous cover	3rd	% WA	0–41.3
Bare soils + herbaceous	3rd	% WA	0.37–60.7
Mine sites or tailings	3rd	Count	0–136
Mine sites or tailings	4th	Count	0–12

Walleye and northern pike population Hg concentrations were calculated at 375 and 675 mm standard lengths, respectively. Each fish population Hg concentrations, normalized at standard length, was calculated from a minimum of 10 fish specimens collected in a lake for a given sampling period. Fish age determination was performed on aging structures, such as opercula, otoliths, and (or) dorsal spines for walleyes, and cleithra or scales for northern pike (Lavigne et al. 2010). When individual fish ages were available, fish growth rates were calculated for both species in each lake for a given sampling season, again with a minimum of 10 specimens for each sampling effort. Growth rates were included in our matrix as the mean age of the fish to reach the standard length.

Table 3 Climate variables and acid deposition

		Years	Units	Range
Climate variables	Annual temperature[a]	1974–2010	°C	−4.20 to 6.66
	Annual temperature[b] (AT3)	1972–2010	°C	−2.74 to 5.58
	Summer temperature[a]	1974–2010	°C	8.93 to 16.78
	Summer temperature[b]	1972–2010	°C	2.25 to 16.48
	Winter temperature[a] (WT)	1974–2010	°C	−12.27 to 1.75
	Winter temperature[b] (WT3)	1972–2010	°C	−12.16 to −0.96
	Annual precipitation[a]	1974–2010	mm	183 to 1187
	Annual precipitation[b]	1972–2010	mm	363 to 1072
	Summer precipitation[a]	1974–2010	mm	13 to 835
	Summer precipitation[b]	1972–2010	mm	76 to 597
	Winter precipitation[a]	1974–2010	mm	18 to 513
	Winter precipitation[b]	1972–2010	mm	119 to 1047
Acid deposition	Annual deposition[a]	1983–2005	$MgSO_4$ m^{-2}	760 to 1775
	Summer deposition[a]	1983–2005	$MgSO_4$ m^{-2}	362 to 1122
	Winter deposition[a]	1983–2005	$MgSO_4$ m^{-2}	285 to 571

Mean summer temperature, precipitation, and acid rain deposition were calculated for the months of May to September inclusively

Mean winter temperature, precipitation and acid rain deposition were calculated for the months of November to April inclusively

[a] Mean for the year of fish catch

[b] Mean for 3 years preceding fish catch

For all 90 lakes included in our matrix, we used a total of 176 free-access Landsat 1, 2, 4, 5, and 7 satellite images with a resolution of 60 m for the Landsat 1 and 2 and with a resolution of 30 m for the Landsat 4, 5, and 7. We also used a rasterized image of the digital elevation model (DEM) of the Canadian digital elevation data (CDED) found on the Geobase Website as well as vectorial topographics maps at a scale of 1:50,000 of the National Topographic Data Base (NTDB) found on the Geogratis Website to determine 15 physiographical variables, to distinguish eight classes of natural vegetation growing on the watershed and to estimate five indices of anthropogenic activities in the watershed (Table 2). We linked biological data and physiographical data by using satellite images taken the same year as that of fish sampling for each lake. Then, only good-quality images taken between June and September were selected. Twenty-four of the 28 variables were obtained at the third-level watershed defined by Beaulne et al. (2012), that is, the land surface directly draining into the lake without transiting through another lake larger than 4 km^2. This choice was justified by the fact that large lakes situated upstream from the studied lake act as retention water bodies and thus greatly minimize the influence of the upstream portion of the watershed on the studied lake. The third-level watershed boundaries were determined using the r.watershed procedure of GRASS ver. 6.4 (Grass Development Team 2011) and map operation using QGIS ver. 1.7 (Quantum GIS Development Team 2011). To do so, we first determined entire watersheds on CDED raster maps and then delimited the third-level watershed with the help of the streams and lakes layer of the NTDB vectorial mapset. The variables "drainage length to the lake" and "drainage density" were obtained after examining the streams and lakes layer of the NTDB maps within the third-level watershed areas, whereas the lake order was defined for the entire watershed area (Table 2). In addition, 2 of the 28 variables were estimated at the fourth watershed level, defined by Beaulne et al. (2012) as the land area within a 1-km buffer zone from the lakeshore (Table 2). We also included measurements of slope steepness of the watershed in the variables that could be deducted from geospatial images as Teisserenc et al. (2011), Moingt et al. (2013), and Beaulne et al. (2012) showed the impact of this physiographical variable on Hg weathering from catchments and on fish Hg concentrations. For this reason, we calculated the mean slope of the third-level watershed and we also distinguished five classes of watershed slopes ranging from 0–2 % to greater than 20 % (SL20) (Table 2).

Even though the 90 studied lakes lie on the Canadian Precambrian Shield, the lowest parts of the region were covered by a very large lake, called the Ojibway–Barlow Lake, upon glaciers retreat approximately 10,000–8000 years ago. After the lake drained, 10- to 60-m thick clay sedimentary deposits, formerly forming the bottom of the lake, were exposed and now constitute the local substratum. Clay deposits is considered as a variable worthy of interest, because clay lakes are more turbid thus influencing the hunting capacity of walleye, which rely on sight to catch their prey (Abrahams and Kattenfeld 1997; Beaulne et al. 2012). We used the geological landscape map produced by Fulton (1995), now found under its digitized form on the Geobase Website, to delimit the clay substratum in the studied region. We then transposed this information as a supplementary layer on the geospatial maps. This allowed us to calculate the percentage of clay substratum (CS) in the third-level watershed for each lake studied (Table 2).

Regarding vegetation cover on the watersheds studied, we identified wetlands and five classes of forests from the geospatial images (Table 2). We also considered the potential impact of two major anthropogenic activities commonly occurring in the watersheds: mining and logging. Doing so, we associated newly observed bare soils and/or herbaceous cover to recent logging activities (three variables in Table 2). This choice is justified by the fact that in the region studied, the natural landscape is mainly composed of closed forest with little rocky outcrops and that small, local, urban settlements or limited agricultural activities have not been significantly expanding over past decades. The number of mine sites and/or tailings, either in the third-level watershed or in the 1-km buffer zone from the lakeshore (two variables in Table 2), was identified on digital maps of the mining area layer of the NTDB.

As far as climate variables are concerned, we considered six temperature variables and six precipitation variables (Table 3). To get these variables, we used Canadian Daily Climate Data (CDCD) from 1976 to 2007 and online data from 2007 to 2010 from on the Environment Canada Weather Office Website for 62 meteorological stations covering the region comprised within 45 and 55°N and −71 and −81°W. For each lake studied, we calculated the annual mean air temperature and precipitation, as well as the means for winter months (November to April inclusively) and the means for summer months (May to September inclusively). Mean temperatures and precipitations were calculated over the lake watershed with a 1-km^2 resolution grid interpolated from the measured values at each meteorological station. The same approach was employed to calculate mean temperatures and precipitations over 3 years prior to the year of fish catching for a given lake. This approach was selected to avoid the impact of singular meteorological events that could mitigate the long-term Hg accumulation trends in fish and add noise on models predictions. To do so, we applied the Inverse Distance Weighted (IDW) interpolation procedure with the v.surf.idw module of GRASS GIS (Neteler and Mitasova 2008) that uses vector maps created from the data points obtained at each meteorological station, for a given year.

In order to get acid deposition data, we used the Canadian Precipitation Monitoring Network (CAPMoN) found on the Environment Canada CAPMoN Website. Throughout Canada, 37 sites have been part of that network and have been sampled daily from July 1984 to the end of December 2007. To get a sufficient number of interpolation points, we used all CAPMoN sampling sites and interpolated the data for the region covered by this study. We applied the IDW interpolation procedure as described above, and we included the mean annual, mean summer, and mean winter acid deposition in our matrix (Table 3).

Estimation of Fish Mercury Concentrations at Standard Lengths

Polynomial regression analysis with indicator variables (Tremblay et al. 1998) was used to describe fish mercury concentrations for each sampled lake and for each sampling year. This statistical approach is best adapted for revealing mercury versus length relationships that are not always linear and thus not suitable for a covariance analysis. Moreover, estimated fish mercury concentrations obtained at standard lengths by polynomial regressions for each fish population are compared at their 95 % confidence intervals. For each lake, each sampling year, and each fish species sampled, individual fish total length was plotted against mercury concentration to fit a curve described by Tremblay et al. (1998) in their equation using nonlinear modeling procedure in JMP ver. 7 statistical package (SAS Institute 2007). The fitted model was used to compute the mean fish mercury concentrations at standard length as well as the 95 % confidence intervals for a given species, in a given lake, in a given sampling year.

Estimation of Fish Age at Standard Lengths

We used the von Bertalanffy growth model (Ricker 1980) to estimate mean ages at standard length of walleye and northern pike populations (375 and 675 mm respectively) for which we had age measurements. We fitted the fish ages versus fish total length plot to the von Bertalanffy growth equation. Using the nonlinear JMP ver. 7 statistical package (SAS Institute 2007) procedure, we estimated the parameters of the inverse prediction, thus providing a fish age at a standard length.

Model Building Process

Prior to any subsequent step, we log10-transformed fish Hg concentrations at standard lengths to get a better fitting against the predictive variables, as the partial relationship is not always linear. Normality and the homoscedasticity of the residuals are easily obtained with this data transformation. In a first step, we used the mixed procedure that alternate forward and backward steps to identify the most significant variables with a probability level at 0.05 and include the best predictive variables as a stepwise variable selection technique (JMP Fit Model process with Stepwise option). Then, a collinearity diagnostic was performed (Condition Index and Variance Inflation Index). The first linear regression models obtained were compared to alternative ones that could be built with the same tools, but starting with different variables that had been excluded in the first models due to their collinearity with other variables. Only the most relevant models were kept, according to the information they bring regarding ways to understand the mechanisms leading to Hg bioaccumulation in fish. We also preferred the models running on the easiest variables that can be collected without tedious and expensive sampling procedures.

Results

Mercury Concentrations and Fish Ages at Standard Lengths

Walleye population Hg levels in 73 Quebec lakes were calculated at 375-mm standard length from the original individual fish Hg concentrations. Those length-normalized Hg concentrations ranged one order of magnitude from 129 to 1255 ng g^{-1} (mean 571 ng g^{-1}, SD 242), and estimated walleye ages to reach the 375-mm standard length spanned fivefold from 2.2 to 10 years (mean 6.7 years, SD 2.0; Table 4). For northern pike populations sampled in 71 lakes, Hg levels calculated at 675 mm standard length also ranged one order of magnitude from 196 to 1738 ng g^{-1} (mean 795 ng g^{-1}, SD 346). The estimated northern pike ages to reach the standard length fell between 3.4 and 8.7 years (mean 6.3 years, SD 1.3; Table 4).

Modeling Walleye Hg Concentrations

We used a stepwise variable selection to generate a multivariate linear regression prediction model for walleye Hg concentrations at 375-mm standard length, including all 44 available variables. Although we had walleye Hg concentrations for 73 lakes, spread over mid-northern Quebec, we only considered 52 series of observations with available walleye growth rates. As a matter of fact, the mean walleye Hg concentration in this subset of 52 lakes (573 ng g^{-1}, SD 232) was very similar to the one found for the entire dataset of 73 lakes. The corresponding adjusted r^2 was as good as 0.79 with a residual standard deviation of 0.08 (Table 5). Estimated fish ages at 375-mm standard length appeared to represent the strongest driving variable, explaining by itself 54 % of the variations of the Hg concentrations (Fig. 1). In other words, lower walleye growth rates definitively corresponded to higher fish Hg concentrations throughout the entire region under study, which is in agreement with what previous studies reported for this particular species (Lavigne et al. 2010; Simoneau et al. 2005).

Using again a stepwise variable selection, we obtained the best prediction model for walleye growth rates, expressed as calculated fish ages at 375-mm standard length, obtained for 52 observations and considering 40 variables (all 44 variables except growth rates and the 3 acid deposition variables). This model yielded an adjusted r^2 of 0.47 with a residual standard deviation of 1.49 (Table 5). Walleye growth rates mainly appeared to be stimulated by warmer mean winter temperatures of the last 3 years prior to fish sampling (WT3). This variable WT3 explained 36 % of the variations of walleye growth rates (Fig. 2).

Modeling Northern Pike Hg Concentrations

Even though our data bank is comprised of 73 lakes with measurements on northern pike, we only had 36 series of observations with pike growth rates. Mean Hg concentration on northern pike in these 36 lakes was slightly higher but not significantly different from that found in the 73 lakes (Test z to compare means, $z = 0.71$ and $p = 0.48$). We thus ran a stepwise variable selection to produce a multivariate regression model for northern pike Hg concentrations at 675-mm standard length (836 ng g^{-1}, SD 384), considering 41 variables (all 44 variables except the 3 acid deposition variables). This model yielded a good adjusted r^2 of 0.70, with a residual standard deviation of 0.13 (Table 5). Pike Hg concentrations firstly appeared inversely related to mean slopes in the third-level watershed (MS), the highest pike Hg concentrations being found in lakes located in the flattest landscapes. The MS variable explained 31 % of the observed variations of pike Hg concentrations (Fig. 3). Pike Hg concentrations also appeared positively correlated to mean annual temperatures calculated over 3 years prior to fish sampling (AT3)

Table 4 Intervals of Hg concentrations and fish ages at standard lengths

		Mean	SD	Min.	Max.
Walleye	Hg L_{375} (ng g^{-1})	571	242	129	1255
	Ages L_{375} (years)	6.7	2.0	2.2	10.0
Northern Pike	Hg L_{675} (ng g^{-1})	795	346	196	1738
	Ages L_{375} (years)	6.3	1.3	3.4	8.7

Table 5 Models predicting fish Hg concentrations and fish ages at standard lengths for Walleye (Eqs. 1 and 2) and Northern Pike (Eqs. 3–5) considering geomorphic variables, vegetation in the watershed, anthropogenic activities, climate variables, and acid deposition

Walleye	(1) $\mathrm{Log_{10}\,Hg\,L_{375}} = 6.23 \pm 0.52 + 0.06 \pm 0.007$ Ages $\mathrm{L_{375}} - 0.08 \pm 0.01$ LAT $- 0.039\ pm0.009$ SL20 $- 0.013 \pm 0.006$ WT $+ 0.008 \pm 0.004$ BS
	$n = 51$, $r^2_{\mathrm{adj}} = 0.79$, RMSE* $= 0.08$
	(2) Ages $\mathrm{L_{375}} = 1.89 \pm 0.82 - 0.34 \pm 0.09$ WT3 $+ 0.10 \pm 0.03$ WB $+ 0.05 \pm 0.02$ CPDS
	$n = 51$, $r^2_{\mathrm{adj}} = 0.47$, RMSE* $= 1.49$
Northern Pike	(3) $\mathrm{Log_{10}Hg\,L_{675}} = 2.50 \pm 0.18 - 0.10 \pm 0.02$ MS $- 0.015 \pm 0.003$ WA/LA $+ 0.13 \pm 0.02$ Ages L675 $+ 0.16 \pm 0.03$ AT3
	$n = 36$, $r^2_{\mathrm{adj}} = 0.70$, RMSE* $= 0.13$
	(4) Ages $\mathrm{L_{675}} = 7.59 \pm 0.20 - 0.43 \pm 0.10$ SL20 $+ -0.08 \pm 0.02$ MF $- 0.69 \pm 0.13$ AT3
	$n = 36$, $r^2_{\mathrm{adj}} = 0.74$, RMSE* $= 0.65$
	(5) Ages $\mathrm{L_{675}} = 5.97 \pm 0.35 + 0.016 \pm 0.004$ CS $- 0.44 \pm 0.13$ SL20
	$n = 36$, $r^2_{\mathrm{adj}} = 0.64$, RMSE* $= 0.77$

The signification of the variables is given in Tables 2 and 3

Parameters values are followed by their standard errors (±standard errors) and the independent variables are listed in order from the higher explanation to the lowest

* Root mean square error

(Fig. 4). The AT3 variable explained 27 % of the observed variations of pike Hg concentrations.

A prediction model also was generated for northern pike growth rates, represented by their ages at 675-mm standard length, considering 43 variables (all 44 variables to the exception of growth rates) for 36 series of observations. The adjusted r^2 of this model was as good as 0.74 with a residual standard deviation of 0.65 (Table 5). The highest northern pike growth rates appeared strongly correlated to the percentage of watershed slopes steeper than 20 % (SL20); this variable explained as much as 62 % of the variations of the growth rates (Fig. 5).

We also generated an alternative linear regression model, still considering all variables with the exception of growth rates for 36 series of observations. In this new model, we selected the variables clay substratum (CS) and the percentage of watershed slopes steeper than 20 % in the third-level watershed (SL20). We obtained an adjusted r^2 of 0.64 with a residual standard deviation of 0.77 (Table 5). In that alternative model, the lowest pike growth rates appeared now strongly related to the percentage of watershed area with clay substratum, with 53 % of the variations of pike growth rates explained by this variable (Fig. 6).

Discussion

Influence of Physiographical Variables on Fish Hg Concentrations

Walleye Hg concentrations are highly influenced by fish growth rates, where faster fish growth rates are related to the lower Hg concentrations (Table 5). Any variable influencing the growth rates of that particular fish species will thus have modulating influence on Hg concentrations. Rather flat landscapes, expressed as exhibiting fewer slopes steeper than 20 % in the third-level watershed (SL20), explained approximately 11 % of variations towards higher Hg concentrations (Table 5). Similarly, the presence of more wetlands adjacent to the sampled lakes, that is, located within a 1-km buffer zone around the lake shores (WB) and a higher fraction of the third-level watershed covered by coniferous forest growing on poorly drained soils (CPDS) are characteristic physiographical variables of flat landscapes. Both WB and CPDS variables were associated with lower walleye growth rates (Table 5), corresponding to higher Hg levels. The observed relationship between flat landscapes and higher Hg concentrations as well as lower growth rates could be explained by the fact that walleye is a pelagic predatory fish species, thriving in cool waters, with definite hunting habits by sight (Lester et al. 2004; Scott and Crossman 1973). In flat

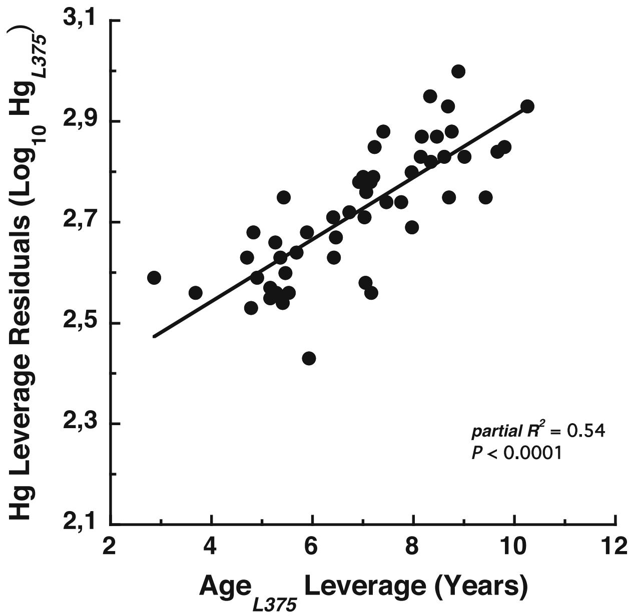

Fig. 1 Walleye mercury concentrations at 375-mm standard length vs. fish age at that standard length

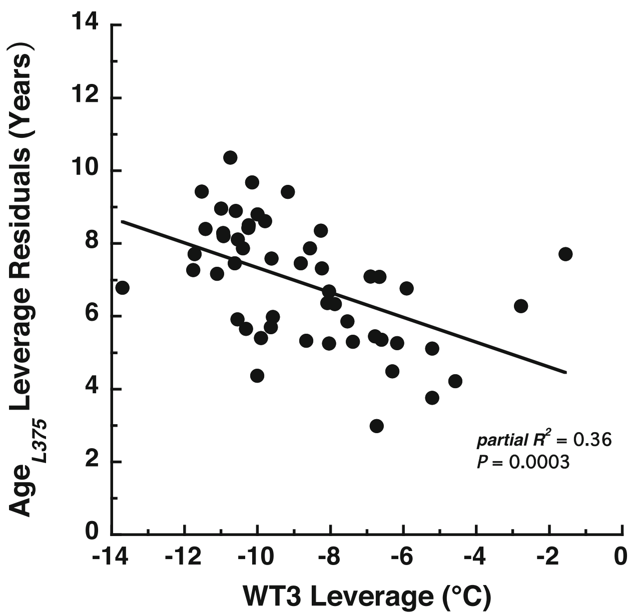

Fig. 2 Walleye ages at 375-mm standard length vs. mean winter temperatures over three consecutive winter seasons prior to fish sampling

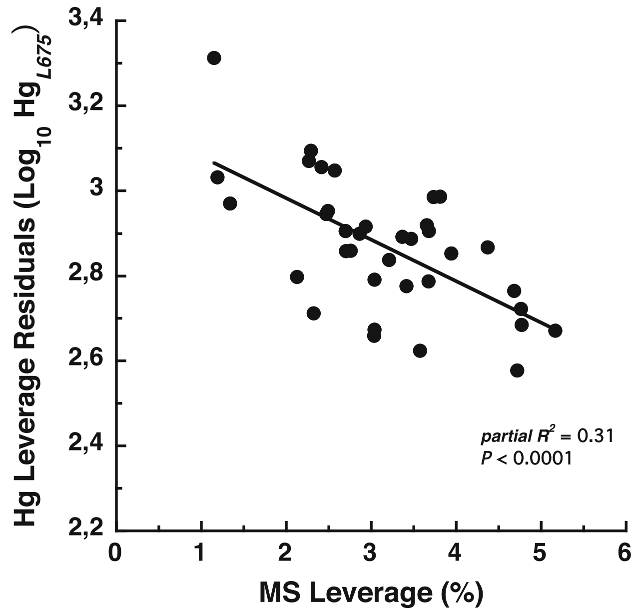

Fig. 3 Pike Hg concentrations at 675-mm standard length vs. mean slope of lake watershed

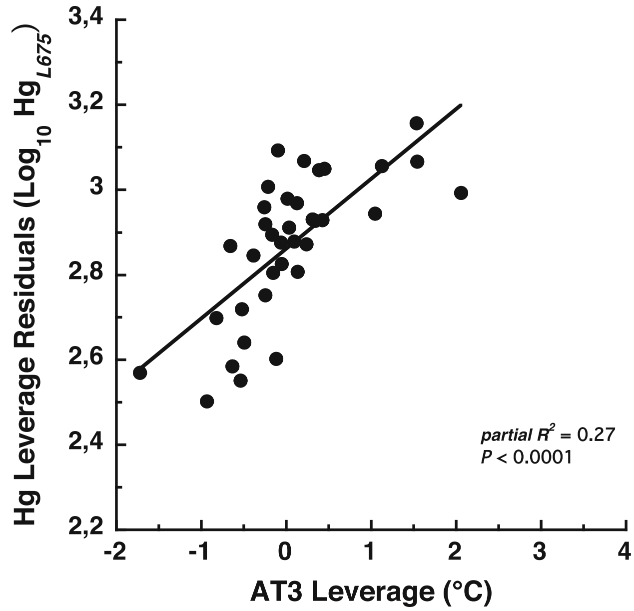

Fig. 4 Pike Hg concentrations at 675-mm standard length vs. mean annual temperatures over 3 years prior to fish sampling

landscapes of mid-northern Quebec, lakes are mostly shallow, warmer, and often rather turbid because of bottom sediment resuspension under wave action. This environmental setting represents non-optimal conditions for walleye, resulting in lower growth rates, and consequently higher Hg concentrations.

Until we considered the influence of physiographical variables on walleye Hg concentrations, the Abitibi region constituted a paradox. Indeed, Abitibi is one of the southernmost regions of the large latitudinal span covered by this study and walleye growth rate were thus expected to be higher because of warmer climate. However, it is a region with some of the highest fish Hg concentrations at 375-mm standard length. This paradox now could be explained by

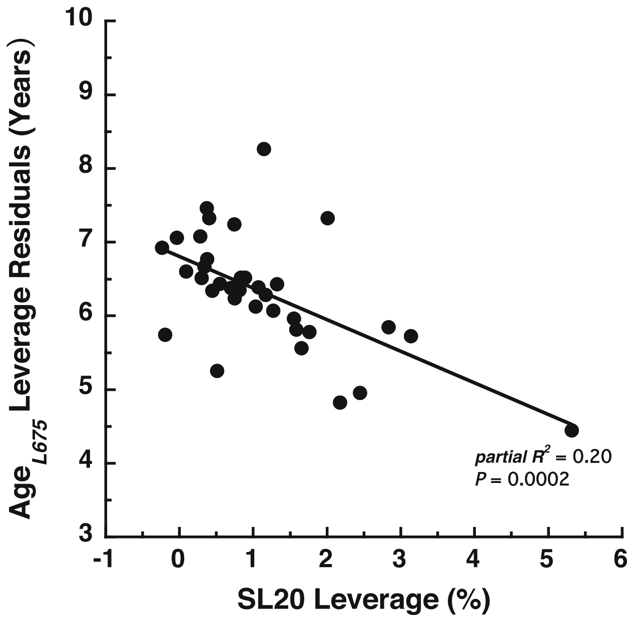

Fig. 5 Pike ages at 675-mm standard length vs. percentage of watershed slopes steeper than 20 %

the fact that this region also is generally characterized by a very flat landscape, an important variable controlling in part walleye Hg concentrations. As such, we observed that latitudes (LAT) were negatively correlated to walleye Hg concentrations, this variable explaining 16 % of the variations (Table 5). The observed negative correlation between walleye Hg concentrations and LAT is probably attributable to the fact that a fair number of walleye in our data bank were collected in the Abitibi region, characterized by a flat landscape, which drove the linear regression model in that unexpected direction.

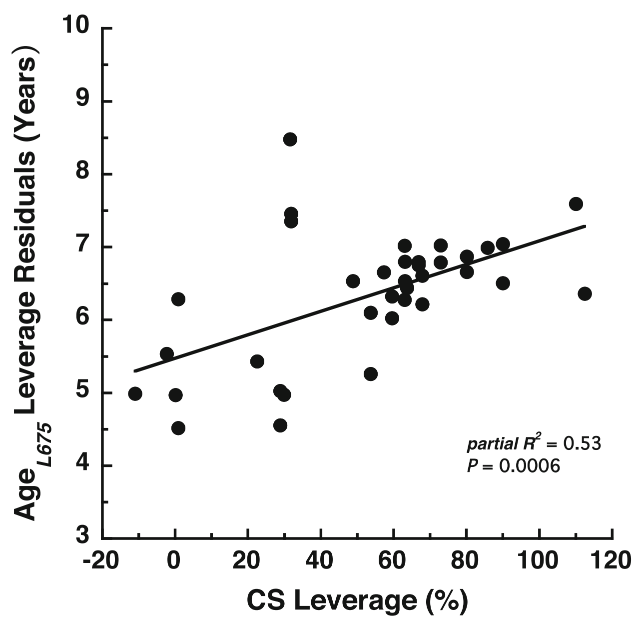

Fig. 6 Pike ages at 675-mm standard length vs. percentage of watershed area with clay substratum

Northern pike is an omnivorous fish species mostly feeding in shallow littoral areas (Scott and Crossman 1973). Shallow littoral zones of lakes located in relatively flat landscapes have been described as receiving high mercury and methyl Hg loadings from nearby wetlands and poorly drained acidic soils (Burns et al. 2012; St. Louis et al. 1994, 1996; Teisserenc et al. 2011). The combination of the latter two observations could explain why northern pike of this study caught in lakes characterized by low values of mean watershed slope (MS) exhibit higher Hg concentrations (Fig. 3). In addition, we also observed that lower pike growth rates are related to smaller fractions of the third-level watershed having steep slopes (SL20) and/or having mixed forest (less than 50 % coniferous stands usually meaning steeper slopes) (MF) as well as higher fractions of clay substratum (corresponding to flat terrains) (CS) (Table 5; Figs. 5 and 6). Because northern pike Hg concentrations were negatively correlated to fish growth rates, although not as strongly as for walleye (Table 5), the combination of these three physiographical variables (SL20, CS, MF) characterizing flat landscapes appeared to correspond to higher Hg concentrations. A similar explanation can be given to interpret the relationship between smaller watershed area to lake area ratios (WA/LA) and higher Hg concentrations (Table 5). Lakes with small WA/LA are usually oligotrophic headwater lakes, thus only sustaining slow northern pike growth rates, which are related to high Hg concentrations. Our finding contradicts the observations of Gantner et al. (2010a, b) for landlocked Arctic char, where higher Hg concentrations were observed under higher WA/LA conditions. However, these authors did not measure fish growth rates to further interpret their data.

Climate Control on Fish Hg Concentrations

Our models show that walleye is a fish species influenced by climate conditions. Higher fish growth rates were notably related to warmer mean temperature over the three winter seasons preceding fish sampling (WT3) (Fig. 2). Harris and Bodaly (1998) reported that walleye increase their metabolic rates in warmer waters. In addition, more shallow thermoclines are expected with climate warming (King et al. 1999), which increases the volume of cool water habitat in which walleye thrive, and consequently may have beneficial growth consequences for that species (Quist et al. 2002). Taking into account the strong relationship between faster growth rates and lower Hg concentrations for walleye (Fig. 1), warmer mean winter temperatures will correspond to lower fish Hg concentrations. Similarly, we observed a direct but weaker relationship between warmer mean winter temperature 1 year prior to fish sampling (WT) and lower Hg concentrations; this relationship explained 3 % of the observed variations of fish Hg concentrations (Table 5). Our results are in agreement with those of French et al. (2006) for two salmon species caught in Lake Ontario between 1983 and 2003, which reported lower Hg concentrations following higher mean annual temperatures in the previous year. The latter authors attributed this relationship to temperature dependent prey fish dynamics at the base of the aquatic food chain. Rennie et al. (2010) also found that warmer conditions during a 4-decade period corresponded to lower Hg concentrations in two species of coregonid fish in Ontario lakes. They explained the latter observation with a decline in fish body condition (i.e., the relative weight of fish specimens with respect to standard weight), but these authors did not include fish ages in their analysis.

Northern pike appeared to be a fish species even more sensitive to climate conditions than walleye. Slowest northern pike growth rates were correlated to coldest annual temperature (mean of 3 years prior to fish sampling, AT3), the latter variable explaining 18 % of the variations in growth rates (Fig. 7). Recalling that there is an inverse relationship between pike growth rates and Hg concentrations (Table 5), although weaker than that of walleye (8 % of the variations explained as compared to 54 % for walleye), one would consequently expect that warmer conditions would also somehow translate into lower pike Hg concentrations, as for walleye. Surprisingly, we observed the contrary: a positive relationship between AT3 and pike Hg concentrations (27 % of the variations explained) (Fig. 4). This observation could be attributed to the fact that northern pike, being a littoral fish, seems more influenced by enhanced Hg methylation conditions in shallow areas under warmer climatic conditions (Bodaly et al. 1993; Ramlal et al. 1993) than by enhanced growth rates. A similar relationship between warming climate and increased Hg concentrations was observed by Carrie et al. (2010) in burbot (*Lota lota*) living in shallow coastal regions of the Mackenzie River in the Arctic.

In an alternative model (not presented), we also found that higher winter precipitation preceding fish sampling explained a few percent of higher Hg concentrations. This observation, similar to that of Rennie et al. (2010) for coregonids, could be explained by higher weathering rates of terrestrial Hg from watersheds (Harris et al. 2007) translating into higher Hg bioaccumulation for that littoral fish species.

No Detectable Influence of Mining on Fish Hg Concentrations

Mid-northern Quebec is a region where Cu–Zn–Au mining is a major economic activity. Of the 90 lakes included in our data set, 49 had large mines and/or tailings in their watersheds. The influence of mining activities on Hg inputs to lakes has been clearly evidenced by high Hg concentrations in recent sediments of the studied region (Petit et al. 2011). Nevertheless, the influence of mining activities could not be detected in any of the regression models that we obtained for walleye or northern pike population Hg concentrations (Table 5). It should first be noted that even in lakes like Chibougamau or Abitibi, with up to 12 and 7 mines and tailings respectively in their 1-km buffer zones around the lake shores, Hg concentrations in the environment are several orders of magnitude lower than those measured in lakes along the Pinchi faultline in British Columbia, a region naturally rich in cinnabar (Petit et al. 2011; Weech et al. 2004). Consequently, Weech et al. (2004) were able to determine the impact of Hg mining on Hg levels in fish from the Pinchi Lake region, contrary to our study. Second, our results corroborate those of Petit et al. (2011) and Moingt et al. (2013) observations, suggesting that Hg accumulating in the sediments of large mid-northern Quebec lakes near mines and/or tailings are not readily methylable and as such hardly enter the aquatic food chain. The large size of the mining lakes that we studied as well as the relatively small accumulation of sedimentary organic matter probably contribute to minimizing sulfate-reducing conditions in surface sediments, which would lead to Hg methylation, contrary to what Winch et al. (2009) observed directly in mine tailings.

On the other hand, we observed a weak positive relationship for northern pike between fish growth rates and the

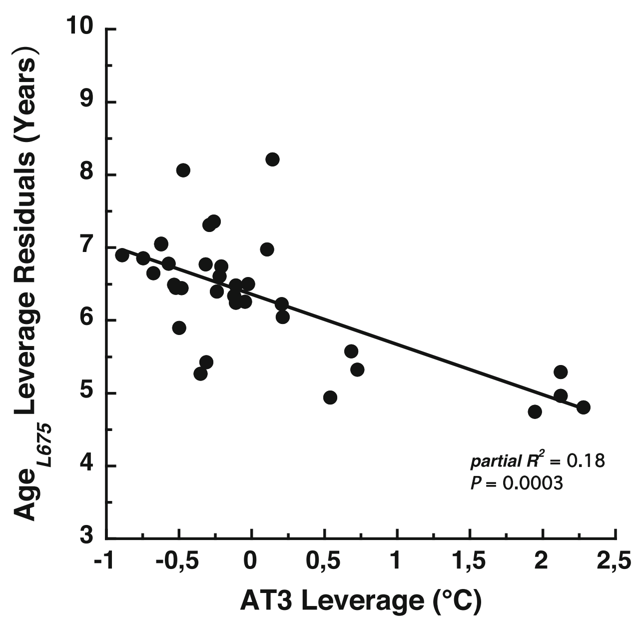

Fig. 7 Pike ages at 675-mm standard length vs. mean annual temperatures over 3 years prior to fish sampling

number of mines and tailings in an alternative model (data not presented). It appears that this surprising relationship was drawn by two points (Chibougamau Lake). Chibougamau Lake being located outside the clay plain, mining activities are probably not contributing to significant enhancement of the water turbidity and as such are not negatively affecting fish growth rates, in particular for walleye. On the other hand, mining activities are possibly triggering higher weathering of terrestrial nutrients, which are then found in coastal lake waters, which in turn enhance northern pike growth rates.

Weak Influence of Industrial Logging on Fish Hg Concentrations

Industrial logging in the boreal forests of Fenno-Scandinavia and Canada have been shown to significantly increase Hg and MeHg fluxes from watersheds to experimental lakes, particularly when runoff waters are naturally characterized with low MeHg and low DOC concentrations (Bishop et al. 2009; Porvari et al. 2003; Sørensen et al. 2009). Garcia and Carignan (1999) and Desrosiers et al. (2006) attributed the increases in Hg concentrations in zooplankton and periphyton in a series of experimental lakes in Quebec impacted by logging activities to the combined effect of increased Hg loadings, more favorable conditions of Hg methylation, and changes in algal communities. Subsequently, Garcia and Carignan (2000, 2005) reported a twofold increase in Hg concentrations in four species of fish (including northern pike and walleye) from 9 logged lakes versus 20 reference lakes. But these observations were obtained on small experimental headwater lakes, with water surface areas comprised between 0.2 and 0.8 km^2. In our data set compiled for large lakes of mid-northern Quebec, we identified recent industrial logging activities by the surface of a given watershed recently showing bare soils and/or herbaceous cover (Table 2). We only found a weak, positive relationship between logging activities and walleye Hg concentrations; the fraction of bare soils in the lake watershed (BS) explaining 2 % of the variations (Table 5). Along with the recent findings of de Wit et al. (2014) reporting that forest management could not be linked to any increase in MeHg in streams, it seems that Hg concentrations in predatory fish of large Quebec lakes were not significantly affected by industrial logging, even in cases where those activities resulted in cutting up to one fifth of the watershed surface over a 10-year period preceding fish sampling. This result differs from those of Sampaio et al. (2009) factorial analysis relating deforestation at the watershed scale in the Amazon to higher Hg concentrations in 10 fish species frequently consumed by the local populations. These authors calculated that fish living downstream of the most deforested watershed of their study (>60 % of its surface area) had on average 20 % more mercury in their flesh than fish living in pristine environments.

No Discernable Influence of Acid Precipitation on Fish Hg Concentrations

Several studies have established a link between acidic waters and higher Hg concentrations in aquatic biota, particularly in fish at the top of the food chain (Hrabik and Watras 2002; Lacoul et al. 2011; Rennie et al. 2005; Scheuhammer and Graham 1999). The biogeochemical processes invoked are either direct, such as higher Hg methylation with lower pH (Graham et al. 2012), or indirect such as slower growth rates of upper-trophic-level organisms under more acidic conditions (Lacoul et al. 2011). Inversely, other authors have reported enhanced Hg concentrations in fish after significant reductions in acid rain deposition (Hongve et al. 2012; Wyn et al. 2010). In our study, the effect of the temporal variations in acid rain deposition on walleye or northern pike Hg concentrations could not be detected with the available data set. This may be attributable to the fact that the Canadian Precipitation Monitoring Network was only maintained by the Government of Canada from July 1984 to the end of December 2007, thus not covering the entire 1976–2010 period during which this study was conducted.

Observing Easily Accessible Natural and Anthropogenic Factors to Predict Game Fish Hg Concentrations

Contrary to the Great Lakes or St Lawrence River region regions (Monson et al. 2011; Neff et al. 2013), the absence of long-term environmental monitoring programs in large natural lakes in Quebec hampers the direct drawing of regional risk maps or temporal trends of Hg concentrations in fish over the past decades. After our extensive fish data mining in Quebec, we found that of the 90 lakes reliably sampled over a latitude span of 7° for determining walleye and northern pike Hg concentrations, very few of them had been sampled twice and three times respectively during the 1976–2010 period (Table 1). By pooling all 44 identified variables for the 90 lakes over three decades, we nonetheless managed to highlight driving factors ultimately interacting to control Hg concentrations in both fish species. Fish growth rate, a variable rarely measured in fish monitoring programs, represents a key variable to interpret spatial and temporal fluctuations in walleye Hg concentrations and to some extent northern pike Hg concentrations. Flat landscapes represented either by weak slopes in the watershed, clay substratum, wetlands, or coniferous stands growing on poorly drained soils then appear to either be directly related to higher Hg concentrations or to correspond to lower fish growth rates, consequently also corresponding to higher Hg concentrations for both fish species. Higher mean winter or annual temperatures appeared to stimulate walleye and northern pike growth rates. Whereas these warmer conditions ultimately corresponded to lower Hg concentrations in walleye, they turned out to be related to higher Hg concentrations in northern pike. These contrasting observations may be explained by the distinct preferred habitats of these fish species, walleye being a pelagic species probably benefiting from shallower thermoclines under warmer conditions, whereas northern pike are a littoral species most likely to be influenced by enhanced Hg methylation conditions in warmer, shallow environments. These results suggest that global climate warming may translate in the short-term to lower or higher Hg concentrations in fish flesh depending upon the species. Finally, our comprehensive study demonstrated that neither acid rain, extensive industrial logging, or mining triggered significant changes over time in Hg concentrations in walleye or northern pike living in large natural lakes of Quebec. This surprising result is probably explained by the highly buffered watershed-lake dynamics of these large ecosystems of the boreal forest domain. Caution should be taken not to generalize these findings to smaller lakes, which may exhibit a more direct relationship between Hg concentrations in fish and human activities in the watersheds.

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