Increasing Mercury in Yellow Perch at a Hotspot in Atlantic Canada, Kejimkujik National Park

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In the mid-1990s, yellow perch (*Perca flavescens*) and common loons (*Gavia immer*) from Kejimkujik National Park and National Historic Site (KNPNHS), Nova Scotia, Canada, had among the highest mercury (Hg) concentrations across North America. In 2006 and 2007, we re-examined 16 lakes to determine whether there have been changes in Hg in the loon's preferred prey, yellow perch. Total Hg concentrations were measured in up to nine perch in each of three size classes (5−10 cm, 10−15 cm, and 15−20 cm) consumed by loons. Between 1996/97 and 2006/07, polynomial regressions indicated that Hg in yellow perch increased an average of 29% in ten lakes, decreased an average of 21% in three, and were unchanged in the remaining three lakes. In 2006/07, perch in 75% of the study lakes had Hg concentrations (standardized to 12-cm fish length) equal to or above the concentration (0.21 $\mu g \cdot g^{-1}$ ww) associated with a 50% reduction in maximum productivity of loons, compared with only 56% of these lakes in 1996/97. Mercury contamination currently poses a greater threat to loon health than a decade ago, and further reductions in anthropogenic emissions should be considered to reduce its impacts on ecosystem health.

Introduction

Mercury (Hg) contamination is an issue of global concern because numerous human activities release Hg into the environment and increase its concentrations in many abiotic and biotic compartments (*1*). These increases are largely the result of Hg emissions from mining, cement production, and the combustion of coal for power generation (*2*). This Hg can be deposited near the source or transported around the globe to contaminate remote environments. Once deposited onto aquatic systems, bacteria rapidly transform inorganic Hg into methylmercury (MeHg), the form that biomagnifies up food webs to potentially toxic levels in top predators (*1, 3*).

Mercury concentrations in fish vary because of among-system differences in their biology and in the chemical and physical characteristics of their habitats, some of which are affected by human activities (*1*). Burning of fossil fuels has acidified freshwaters (*4*) and increased concentrations of Hg in lakes and rivers, both of which are linked to higher Hg in fishes (*3, 5*). Sulfate is a key substrate in the methylation of Hg by bacteria (*6*), and recent reductions in its release and deposition, combined with some controls on local Hg emissions from incinerators, contributed to lower Hg in fishes (up to 5.1% y^{-1}; see for example ref *7*).

Although concentrations of Hg in the atmosphere and precipitation recently decreased near some urban or industrial centers (*8*), global anthropogenic Hg emissions are projected to increase if there are no further restrictions on its use and release (*2*). The objective of this study was to examine whether total Hg (THg) concentrations in fish decreased in a region of Atlantic Canada known to be a biological Hg hotspot, Kejimkujik National Park and National Historic Site (KNPNHS) (*9*). Studies in the mid-1990s showed that the common loons (*Gavia immer*) in KNPNHS had two to six times more Hg in their blood than loons in other parts of North America (*10, 11*). Yellow perch (*Perca flavescens*) are the preferred prey of loons (*12*), and a majority of the KNPNHS lakes examined in 1996 and 1997 contained perch with mean THg concentrations exceeding the 0.21 $\mu g \cdot g^{-1}$ (wet weight, ww) threshold associated with a 50% reduction in loon maximum productivity (*13, 14*). Although concentrations of Hg in precipitation declined by 2.0% yr^{-1} at KNPNHS, total wet deposition of Hg has not changed appreciably and concentrations of total gaseous mercury (TGM) at this site are increasing [∼0.3% yr^{-1} (*8, 15*)]. In 2006 and 2007, we revisited these acidic lakes to determine whether THg concentrations in yellow perch changed over the past decade and to relate any changes to physical or chemical characteristics of the lakes or biological characteristics of the fish.

Methods

Study Site. KNPNHS is in southwestern Nova Scotia, Canada (Supporting Information, Figure S1) and is not impacted by local industrial developments or other point sources of pollutants (*16*). The park is downwind of major North American urban and industrial centers, and Hg and acidifying substances originating from these areas are transported to and deposited in the region (*8, 13*). The lakes are all oligotrophic, polymictic, and acidic (pH < 6) but vary in size (from 24 to 2632 ha), total organic carbon (TOC) content (2.6 to 15.4 $mg \cdot L^{-1}$), and abundance of wetlands (0−35% of drainage basin; Tables S1 and S2, Supporting Information).

Sample Collection. As much as possible, sampling in 2006/07 was done in a manner similar to that of the 1996/97 study (*13*). Fishing occurred in August and September 2006/07 (compared to July and August 1996/97). In both studies, minnow traps, trap nets, and angling were used to catch fish; fyke nets were also used in 2006/07. In each lake, the aim of both studies was to capture nine yellow perch from each size class consumed by common loons: 5−10 cm, 10−15 cm, and 15−20 cm (*12*). Fork length (±0.1 cm), weight (±0.01 g), and scales (for aging) were obtained from each fish; all fish were kept cool on ice and then frozen within 24 h of capture. Fish body condition was calculated as 100 × [weight (g)/length $(cm)^3$]. Tissues were collected and processed for Hg and stable nitrogen isotopes ($\delta^{15}N$, used to approximate trophic position) as described in the Supporting Information and in Wyn et al. (*17*). $\delta^{15}N$ was measured in each fish collected in 2006/07 and in a maximum of four composites per lake for those

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collected in 1996/97. Trichoptera (Limnephilidae) or aquatic Lepidoptera were collected in 2006/07 to determine whether baseline δ^{15}N varied across lakes. Quality assurance information is provided in the Supporting Information.

Surface water samples (n = 1/lake/date) have been collected by helicopter in spring (May/June) and fall (September/October) from a number of lakes in KNPNHS since 1983 as part of an Environment Canada long-term monitoring program (*4*). Each sample was analyzed for pH, conductivity, total nitrogen (TN), total organic carbon (TOC), sulfate, and a suite of metals that periodically included THg (methods described in Vaidya et al. (*18*)). Data presented are means from 1995−1997 and 2005−2007 (n = 6; 3 years of semiannual sampling); exceptions are detailed in Table S2.

Mercury Analyses. Freeze-dried whole body homogenates of individual perch collected in 2006/07 were analyzed for THg using a Milestone DMA-80 at the University of New Brunswick (UNB). Data were converted to wet weight using individual moisture contents. Mean recoveries of certified reference materials were 94.6 ± 2.0% [DORM-2 (dogfish muscle), National Research Council (NRC), Ottawa, ON; 90.3−101.2%, n = 60] in 2006 and 101.9 ± 8.0% [DORM-2 and TORT-2 (lobster hepatopancreas), NRC; 83.8−121.7%, n = 51] in 2007. Precision of replicate samples was 5.9 ± 4.1% (n = 84 triplicates) in 2006 and 1.1 ± 7.8% (n = 38 duplicates) in 2007. Blanks had a mean concentration of 0.01 ± 0.01 $\mu g \cdot g^{-1}$ dw (n = 132); sample results were not corrected for blank values. Additional quality control procedures and results are described in the Supporting Information. Yellow perch collected in 1996/97 were pooled within lake according to size, and then wet, whole body homogenates were analyzed for THg using the methods and quality control procedures detailed in the Supporting Information.

Data Analyses. All data were inspected for normality and homogeneity of variances using the Kolmogorov−Smirnov Lilliefors test and F-ratio, respectively. When necessary, data were $\log_{10}$ (THg, length, weight, conductivity, TOC, TN, SO_4, Cl, Fe) or square-root (Al, Ca, Na, Mn) transformed to approximate normality. Residuals were examined for all analyses, and outliers of individual fish within lakes and dates or lakes within dates were identified as those with Studentized residuals exceeding an absolute value of 3 (*19*). All analyses were performed using SYSTAT 10 for Windows (SYSTAT Software Inc., Chicago, IL) with $\alpha = 0.05$. Fish collected in 1996/97 were pooled prior to THg analysis; therefore, 2006/07 data were mathematically pooled in the same way (means of two to three fish of similar length within each size class in each lake) before the temporal comparison.

Between-year, within lake differences in mean log-lengths, log-weights, and ages of yellow perch were evaluated using two-sample t-tests while temporal changes in δ^{15}N were assessed qualitatively due to low sample sizes and the lack of baseline δ^{15}N for 1996/97. Differences in body condition and growth rates of perch were evaluated using analysis of covariance (ANCOVA) with log-length as the dependent variable and log-weight or age as the covariate, respectively. Temporal changes in THg concentrations were analyzed using polynomial regressions within each lake (*20*). The model was log-THg = LC + LC^2 + year + year*LC + year*LC^2 (where LC = fish length centered); backward stepwise regression was used to identify terms significantly related to log-THg in each lake (see Supporting Information).

Temporal trends in water chemistry (for the periods 1995−1997 and 2005−2007) were evaluated using two sample t-tests. For each variable that changed through time, % changes were regressed against the physical characteristics of the lakes (Table S1) to evaluate whether any characteristics explained these differences.

Mean length of yellow perch across all lakes was 12 cm in both 1996/97 and 2006/07, and THg concentrations were standardized to this length to facilitate comparisons. Standardized THg concentrations were calculated by applying the centered log-length for each lake (i.e, log-12 cm − mean log-length$_{lake,year}$) to the lake-specific polynomial regression equations (*21*). Size-standardized log-THg in yellow perch were compared to water chemistry parameters [Table S2, plus Cl, Fe, K, Mg, and Mn (data not shown)] by simple regressions (general linear model, GLM) across all lakes sampled in 1996/97 or in 2006/07 (using pooled data). Both absolute and % change [calculated as (2006/07)/(1996/97) × 100] of perch THg were regressed against physical characteristics of the lakes or against % change of biological or chemical variables that showed significant differences through time (simple GLM). Multiple stepwise regression (retaining variables with F statistic >4 and eliminating collinearity) was also performed across lakes to find the set of variables that best described % change of THg in perch.

Results

Significant increases in THg concentrations of yellow perch were recorded for many lakes in KNPNHS over the past decade (Table 1). Between 1996/97 and 2006/07, mean THg concentrations increased between 10.5 and 58.3% (polynomial regressions, $p < 0.004$) in yellow perch from ten lakes (two were reduced to nonsignificance when one outlier was removed from each lake). For the remaining six lakes, THg in perch decreased (11.1 − 36.8%, $p < 0.001$) in three or did not change (see Table S3 and Figure S2, Supporting Information, for polynomial regressions). Increases in THg occurred predominantly in the 5−10 and 10−15 cm-sized fish, with few increases recorded for the 15−20 cm perch (Figure 1; raw data shown in Figure S3, Supporting Information).

Mean log-length and log-weight did not differ significantly ($p > 0.37$) within each lake from 1996/97 (overall means of 11.9 ± 3.6 cm and 25.79 ± 22.00 g, respectively) to 2006/07 (12.2 ± 3.4 cm and 26.50 ± 21.56 g), but there were some differences in fish age, condition, growth, and δ^{15}N (Table 1; Tables S4 and S5, Supporting Information). Compared to fish captured in 1996/97, yellow perch from 2006/07 were significantly younger in two lakes (two-sample t-test, $p < 0.03$) and had lower condition in eight lakes (intercept of ANCOVA, $p < 0.07$; Table 1, plus Kejimkujik, Pebbleloggitch, and Peskowesk lakes). Growth rates of yellow perch increased between these two periods in three lakes (ANCOVA interaction, $p < 0.04$) but decreased in another two lakes ($p < 0.01$; Table S4). A qualitative analysis of δ^{15}N also suggested that perch in seven lakes were at a higher trophic position in 2006/07 than in 1996/97 (assuming no change in basal δ^{15}N; Table S5). Overall, of the ten lakes with increases in perch THg, six had decreases in condition, three had decreases in age, three had increases in growth, and five had increases in δ^{15}N.

Changes in water chemistry were observed for many lakes in KNPNHS between 1995−97 and 2005−07. TN concentrations in the surface water increased in all 16 lakes by 5 to 15 $mg \cdot L^{-1}$ ($p < 0.07$; Table S2). Aqueous sulfate concentrations decreased by 0.1 to 0.3 $mg \cdot L^{-1}$ in most lakes, with significant reductions in six lakes ($p < 0.04$; Table S2). Significant increases in pH (by 0.1 to 0.2 units) were measured in three lakes ($p < 0.05$; Table S2). Alkalinity, conductivity, TOC, Al, Ca, Fe, Mg, K, Na, and Cl were statistically similar ($p > 0.19$) in 1995−1997 and 2005−2007 for the 16 lakes (insufficient data available to test THg), although small increases in TOC, THg, alkalinity, and conductivity were noted for many lakes between the two periods (Table S2).

The changes in THg in perch were explained by very few factors. For all lakes, the absolute and % change of the length-standardized THg in perch were not related to any physical

TABLE 1. Mean (± SD) Body Condition, Age, and Total Mercury Concentrations (THg, raw and standardized) of Yellow Perch Caught in 1996/97 (*13*) and 2006/07[c] from 16 Lakes in Kejimkujik[a,b]

lake	year	*n*	condition (g·cm^{-3})	age (y)	THg (μg·g^{-1} ww)	12-cm THg (μg·g^{-1} ww)
Back	1996	9	1.11 ± 0.08a	5.0 ± 2.1	0.13 ± 0.06a	0.12
	2006	10	1.04 ± 0.08**	3.6 ± 1.1	0.21 ± 0.04	0.19***
Beaverskin	1996	10	1.10 ± 0.05a	3.7 ± 1.3	0.19 ± 0.08abc	0.20
	2006	8	1.11 ± 0.06	4.2 ± 1.9	0.29 ± 0.02	0.28**
Big Dam East	1996	8	1.17 ± 0.08a	4.6 ± 1.9	0.18 ± 0.08abc	0.18
	2006	8	1.06 ± 0.04***	3.9 ± 0.9	0.20 ± 0.06	0.25*
Big Dam West	1996	7	1.31 ± 0.07b	5.4 ± 2.4	0.21 ± 0.10 abc	0.27
	2006	8	1.25 ± 0.06*	4.8 ± 2.1	0.21 ± 0.06	0.24*
Big Red	1996	6	1.20 ± 0.13a	6.8 ± 2.1	0.53 ± 0.14abc	0.41
	2006	7	1.19 ± 0.07	5.8 ± 1.2	0.47 ± 0.12	0.41
Cobrielle	1996	6	1.16 ± 0.07a	5.2 ± 1.2	0.26 ± 0.07abc	0.20
	2006	6	1.10 ± 0.07	4.7 ± 0.8	0.33 ± 0.07	0.25*
Frozen Ocean	1996	9	1.30 ± 0.12b	5.8 ± 2.3	0.26 ± 0.16abc	0.24
	2006	9	1.20 ± 0.08**	4.8 ± 2.4	0.26 ± 0.11	0.22
Kejimkujik	1996	23	1.16 ± 0.09a	5.2 ± 2.3	0.28 ± 0.10bc	0.30
	2006	26	1.21 ± 0.14	5.3 ± 2.3	0.32 ± 0.14	0.29
Loon	1996	9	1.20 ± 0.05a	6.1 ± 2.0	0.26 ± 0.11abc	0.22
	2006	9	1.16 ± 0.09	3.7 ± 1.8*	0.26 ± 0.11	0.30*
Mountain	1996	8	1.17 ± 0.07a	5.1 ± 2.5	0.22 ± 0.12abc	0.19
	2006	9	1.09 ± 0.07*	2.7 ± 1.7*	0.21 ± 0.11	0.21* (0.20)[d]
North	1996	9	1.09 ± 0.06a	5.4 ± 2.2	0.37 ± 0.14c	0.30
Cranberry	2006	9	1.10 ± 0.10	5.6 ± 2.5	0.49 ± 0.24	0.36*
Pebbleloggitch	1996	9	1.16 ± 0.09a	5.2 ± 2.2	0.16 ± 0.04ab	0.19
	2006	9	1.14 ± 0.04	4.3 ± 1.2	0.20 ± 0.05	0.22* (0.19)[d]
Peskawa	1996	20	1.18 ± 0.08a	4.4 ± 2.0	0.24 ± 0.13abc	0.28
	2006	27	1.20 ± 0.13	5.2 ± 2.6	0.25 ± 0.11	0.24*
Peskowesk	1996	8	1.13 ± 0.06a	5.1 ± 2.1	0.28 ± 0.16abc	0.21
	2006	10	1.16 ± 0.08	5.0 ± 2.4	0.36 ± 0.17	0.27*
Puzzle	1996	8	1.08 ± 0.06a	4.9 ± 2.4	0.23 ± 0.10abc	0.25
	2006	9	1.08 ± 0.10	5.5 ± 2.6	0.33 ± 0.12	0.29**
Upper Silver	1996	7	1.08 ± 0.08a	3.7 ± 1.5	0.14 ± 0.06a	0.19
	2006	10	1.08 ± 0.06	2.5 ± 1.7	0.13 ± 0.04	0.12*

[a] Asterisks represent statistical within-lake differences (* $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, *t*-tests, ANCOVA, or polynomial regression) between years. [b] Letters represent statistical differences (ANOVA) among lakes within each period. [c] 2006/07 data mathematically pooled except in Kejimkujik and Peskawa lakes; see Methods for details. [d] Values in brackets had outliers removed.

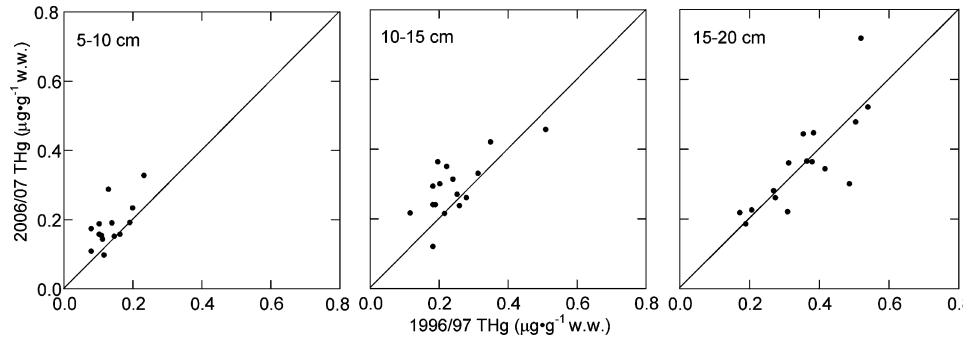

FIGURE 1. Mean THg concentrations of three size classes (5−10, 10−15, and 15−20 cm) of yellow perch captured in 16 lakes in Kejimkujik in 1996/97 and 2006/07. The 1:1 line is shown.

characteristic or to any biological or chemical variable that differed significantly over time (i.e., % change in age, condition, aqueous pH, TN, sulfate; GLM, $p > 0.12$). After Upper Silver Lake was removed as an outlier, the % increase in THg in yellow perch was greatest in lakes with higher pH (using absolute values for 2005−2007; $p = 0.01$, $r^2 = 0.39$; Figure 2a) or lower aqueous THg ($p = 0.01$, $r^2 = 0.40$), TOC ($p = 0.01$, $r^2 = 0.39$; Figure 2b), conductivity ($p = 0.02$, $r^2 = 0.37$), or Al ($p = 0.005$, $r^2 = 0.45$). It must be noted that aqueous THg, TOC, conductivity, and Al were all positively correlated ($r > 0.79$), while also being negatively correlated to pH ($r < -0.78$). The best stepwise model was: % change THg = 9637−1.11 (2005−2007 conductivity) − 147.3(latitude) ($p = 0.08$, $r^2 = 0.34$). Small fish (<10 cm) from the five lakes with the largest % change in THg also showed a significant decline in condition ($p = 0.026$) that was not present in the other lakes or in the larger fish (>15 cm, $p = 0.55$; Kruskal−Wallis).

Discussion

Between 1996/97 and 2006/07, THg concentrations in yellow perch in KNPNHS, a biological Hg hotspot in northeastern North America (*9*), increased an average of 29% in ten lakes, decreased an average of 21% in three lakes, and remained unchanged in the other three lakes. These results were unexpected considering that Hg emissions from North America declined between 1995 and 2000 (*22, 23*), sulfate deposition and Hg concentrations in precipitation decreased in the region (*4, 8*), and total wet Hg deposition to KNPNHS did not change over the same time period (*15*). The change in THg in yellow perch was highest in lakes where fish condition or age decreased, or growth or δ^{15}N increased. Percent change of THg in perch over the decade was also highest in lakes with higher pH or lower aqueous THg, TOC,

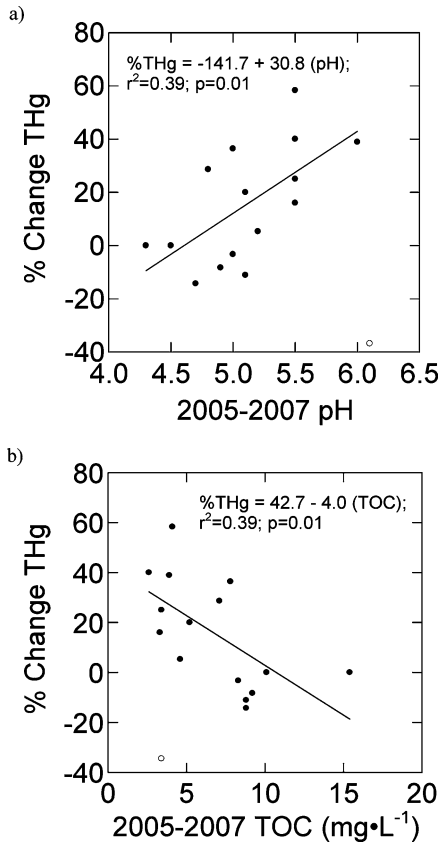

FIGURE 2. Percent change (2006/2007 compared to 1996/1997) of standardized THg concentrations in yellow perch captured in 15 lakes in Kejimkujik versus (a) pH; (b) TOC of the lakes as measured in 2005−2007 (open symbol indicates an outlier, Upper Silver Lake, excluded from the regressions).

conductivity, and Al (as measured in 2005−2007), and the increases occurred primarily in smaller perch (5−15 cm).

The mean annual increase in perch THg concentrations (2.9%) for the 10 lakes in KNPNHS was three times higher than the annual increase (0.8%) for fish in southern Wisconsin, one of the few other North American studies showing recent Hg increases (*24*). The rate of increase at KNPNHS was also similar to some lakes from another study in northeastern North America, where yellow perch THg increased 2.2% y^{-1} (in 6 of 25 lakes) in systems with large watersheds and declining pH and fish condition (*25*). In some lakes in KNPNHS, Hg in yellow perch decreased (2.1% y^{-1}) or did not change over the past decade, a trend that has also been seen in other studies (e.g., ref *7*). Mercury concentrations decreased by 0.5−5.1% y^{-1} in walleye and yellow perch in northern Wisconsin (*7, 24*) and by 0 − 4.1% y^{-1} in yellow perch from the majority of lakes (12 of 25 lakes) in the Adirondacks during the past 10 to 20 years (*25*).

External Influences on THg in Fish. Increases in fish Hg concentrations in other regions were attributed to local increases in wet deposition of Hg or to higher catchment inputs of Hg and acidifying substances (*24, 25*); however, these sources do not appear to have changed for KNPNHS lakes. First, Hg concentrations in precipitation have declined but its total aerial deposition in rainfall was similar over the past decade at KNPNHS (*15*). The precipitation and TGM data for KNPNHS are also comparable to other long-term monitoring sites in the region (*8, 15*). It is possible, however, that other unmeasured sources of Hg to the lakes have increased, e.g., reactive gaseous Hg (RGM). Concentrations of atmospheric TGM increased by ~0.3% yr^{-1} between 1996 and 2004 at KNPNHS (*8*), and the highly water-soluble RGM contributes a minor portion to this atmospheric Hg pool (*26*). Though not measured at KNPNHS, fluxes of RGM to these lakes may have increased at the same time as the increase in TGM. Second, while the release of stored acids and Hg from catchments (especially wetlands) can enhance lake acidities and aqueous Hg and delay recovery of freshwater ecosystems (*1, 3*), lake pH and TOC have not changed significantly over time at KNPNHS (except for pH increases in 3 of 16 lakes). Aqueous THg increased in some lakes, but the data are limited (Table S2) and may not always indicate increases in aqueous MeHg (*25*).

Influence of Chemical Characteristics of Lakes on THg in Fish. Mercury concentrations in fish are typically higher in lakes with lower pH, and more organic carbon and Hg in the water (*1*); indeed, across lakes in KNPNHS in 1996/97, size-standardized THg in perch was higher in lakes with greater aqueous TOC or THg (as well as % wetlands in their watersheds) and lower pH (Figure S4, Supporting Information). However, THg in fish was not related to lake TOC in 2006/07 (Figure S4) and, between 1996/97 and 2006/07, the greatest increases in perch THg concentrations occurred, surprisingly, in lakes with the lowest aqueous concentrations of TOC and highest pH (Figure 2). Although there were some slight, but nonsignificant, increases in THg and TOC in the lakes over time (Table S2), THg data were limited and these changes did not consistently translate into higher Hg in fish. These trends suggest that increased availability, rather than just overall aqueous supply, of inorganic Hg to methylating bacteria and higher concentrations of MeHg at the base of the food web are contributing to the higher Hg in these fish (*1, 17*).

Though not well understood, Hg complexes with organic and inorganic ligands in surface waters and sediments, and its partitioning is determined by the types and characteristics of the ligands and other chemical factors such as conductivity (*27*). The form of Hg determines, in part, its methylation rates because small, neutral molecules are rapidly taken up by methylating bacteria (*28*). Though speculative, the changes in water chemistry seen here may have increased Hg availability to methylating bacteria by changing binding to ligands or the type of dominant complexes present (see for example ref *29*), or rates of methylation using substrates such as sulfate (*6, 28*).

Lakes with high productivity typically have low biotic Hg because of dilution of Hg at the base of the food web (*30*). In KNPNHS, increases in perch THg concentrations were observed in lakes with the lowest productivity, as indicated by conductivity (2005−07). No temporal changes in conductivity were found (other productivity measures not available), but TN increased significantly (2-fold) in all lakes that also had increases in fish Hg (Table S2). It is possible that the higher TN in the most oligotrophic lakes increased MeHg at the base of these food webs, but a recent study also found that nitrate inhibits Hg methylation by bacteria (*31*). Although MeHg data for primary consumers are available for 2006/07 (*17*), neither water (either time period) nor invertebrate (1996/97) MeHg data are available to evaluate temporal trends at the base of these food webs.

Increases in temperature or decreases in oxygen through time may have also caused the observed increases in THg in yellow perch through enhanced Hg methylation and uptake at the base of the food web (*28, 32*). Although mean annual air temperatures at KNPNHS increased from 7.1 and 7.0 °C in 1995 and 1996, respectively, to 7.9 and 8.5 °C in 2005 and 2006, respectively (*33*), recent studies of the 2 deepest lakes in KNPNHS (Kejimkujik and Peskowesk) showed that summer temperature profiles have not changed since the 1970s; however, hypolimnetic oxygen concentrations were significantly lower than previously reported (*34*). Lower oxygen can enhance Hg methylation activities of anaerobic sulfate-

reducing bacteria and could explain the observed increases in fish Hg concentrations (*28*).

Influence of Biological Characteristics on THg in Fish. Decreased fish condition has been linked to increases in their THg concentrations (*25*), and, indeed, condition was reduced in eight KNPNHS lakes, six of which had increases in yellow perch THg between 1996/97 and 2006/07. Increases in size or age, or decreases in growth, could also have caused increases in fish Hg through time (*35*); however, length and weight did not change through time, significant reductions in age were found in three lakes with THg increases, and growth only declined in lakes where there were decreases or no change in fish THg.

Temporal changes in THg occurred primarily in the small (5−15 cm), young (<7 y; Figures 1 and Figure S3) yellow perch. The smallest size class also showed declines in condition between 1996/97 and 2006/07, and these changes were greatest in lakes with the largest increase in THg concentrations. At this time, we cannot explain the changes in condition, but they may be related to declines in the quality or quantity of available prey (*35*) or possibly to Hg toxicity as perch concentrations approach or exceed those known to cause effects in other fishes (*36*).

Implications. The observed increases in THg concentrations in yellow perch in KNPNHS over the past decade suggest that other species in the park are also at greater risk of Hg toxicity. Yellow perch are the preferred prey of common loons (*12*), and loons consuming yellow perch with mean Hg concentrations ≥0.21 $\mu g \cdot g^{-1}$ ww exhibit a 50% reduction in maximum productivity (*14*). In 1996/97, nine lakes had yellow perch with 12-cm mean THg at or above the threshold, but in 2006/07, standardized mean THg concentrations ≥0.21 $\mu g \cdot g^{-1}$ occurred in 12 lakes (14 including outliers; Table 1). In both 1996/97 and 2006/07, yellow perch in Big Red Lake also had a standardized THg concentration of 0.41 $\mu g \cdot g^{-1}$ ww (Table 1), the level for complete reproductive failure in loons (*14*). The increase in the number of lakes exceeding the 0.21 $\mu g \cdot g^{-1}$ threshold and nearing the 0.41 $\mu g \cdot g^{-1}$ threshold suggests that loons in the park will experience more reductions in reproductive success and could eventually exhibit complete reproductive failure on certain lakes if these trends continue (*14*). For example, THg concentrations in yellow perch in North Cranberry Lake could exceed 0.41 $\mu g \cdot g^{-1}$ ww in 2013.

Overall, between 1996/97 and 2006/07, yellow perch THg concentrations increased in more than 60% of lakes examined in KNPNHS at a time when wet deposition and catchment inputs of THg or TOC have not changed; these increases were greatest in the smallest fish and in populations in lakes with the highest pH and lowest THg and TOC. Although the cause is currently speculative, the following factors may have played a role: (1) increased Hg fluxes to the lakes as a result of higher atmospheric concentrations of RGM; (2) increased MeHg production because of higher availability of the inorganic Hg pool to methylating bacteria or of conditions more favorable for methylation (e.g., warmer temperatures, increased TN); and/or (3) reduced quality or quantity of prey available to perch. This study complements only a few others that have shown increases in THg concentrations in fish despite some reductions in emissions of Hg and sulfate in North America. As such, these increases suggest that further reductions in Hg and acidifying emissions from major sources, such as coal-fired generating stations (*2*), should be considered to protect or restore ecosystem health in remote regions such as this one, that were previously identified as Hg hotspots (*9*).

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Supporting Information Available

Details of the sampling, analytical, and statistical methods, and results of the within-year trends in Hg concentrations in perch, are provided. Tables S1 and S2 describe physical and chemical characteristics of the lakes, Table S3 summarizes the polynomial regressions, Tables S4, S5, and S6 describe growth rates, $\delta^{15}N$, and Hg bioaccumulation rates of the perch, and Table S7 summarizes the raw (unpooled) biometric and perch Hg data from 2006/07. Figure S1 is a map of the region, Figures S2 and S3 illustrate the perch THg−length relationships in each lake and each size class, respectively, and Figure S4 shows the relationships between 12-cm Hg and TOC or pH across lakes. This information is available free of charge via the Internet at http://pubs.acs.org.

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