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Factors influencing changes in mercury concentrations in lake water and yellow perch (*Perca flavescens*) in Adirondack lakes

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Abstract Over the past 20+ years, fish with elevated concentrations of mercury (Hg) have been observed in remote lake districts, including the Adirondack region of New York. Across eastern North America studies have also reported a negative correlation between fish Hg concentration and lake pH. Recent controls in emissions of sulfur dioxide (SO_2) have resulted in some improvement in the acid–base status of acid-impacted surface waters including Adirondack lakes. In addition, there has been an apparent decrease in atmospheric Hg deposition. A synoptic survey of 25 lakes in the Adirondacks was conducted in 1992–1993 to analyze spatial patterns of Hg in the water column and yellow perch (*Perca flavescens*). The same cluster of 25 lakes was resurveyed in 2005–2006 to evaluate if changes in lake concentrations of Hg species or fish Hg have occurred. We observed a varied response of changes in water chemistry and fish Hg concentrations. In twelve of the resurveyed lakes the yellow perch had lower Hg concentrations, six lakes had yellow perch with higher Hg concentrations, and in seven lakes yellow perch Hg concentrations did not change significantly ($\alpha = 0.05$). Four variables appear to influence the change in yellow perch Hg concentrations in the Adirondacks: watershed area, elevation, change in pH, and change in fish body condition. We hypothesize that as the acidity in lakes is attenuated, the lakes may become more productive and/or water quality conditions less stressful to fish leading to increasing fish body condition. As fish body condition improves, fish exhibit "growth dilution" of tissue contaminants leading to lower fish Hg concentrations.

Keywords Adirondack lakes · Mercury · Yellow perch · Bioaccumulation

Introduction

Mercury (Hg) contamination is a critical environmental and public health issue for natural resource managers and policy makers. Mercury is a potent neurotoxin that poses a health risk to both humans and wildlife (Driscoll et al. 2007a). Globally atmospheric Hg deposition is approximately three times greater than pre-anthropogenic levels (Mason et al. 1994; Lindberg et al. 2007), with some regions showing increased deposition beyond these levels. The Adirondack region of New York has experienced an approximately sixfold increase in Hg deposition over the past 100 years (Lorey and Driscoll 1999) followed by decreases of 25–30% over the past few decades. Most Hg pollution originates from atmospheric emissions (Fitzgerald et al. 1998) as a result of the combustion of fossil fuels (e.g., electric utilities), industrial processes, and

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incinerators (Driscoll et al. 2007a). Once Hg enters the environment, it may be converted to the more bioavailable form, methylmercury (MeHg). Fish accumulate most of their MeHg burden from food (Hall et al. 1997; Bodaly and Fudge 1999). In the United States, consumption of fish is the primary pathway for exposure of humans to MeHg (Driscoll et al. 2007a).

Over the last two decades, fish with elevated concentrations of Hg have been observed in remote lakes (Grieb et al. 1990; Suns and Hitchin 1990; Bodaly et al. 1993), including the Adirondack region of New York (Driscoll et al. 1994, 1995). Recently, Evers et al. (2007) found five biological Hg “hotspots” in the Northeast, including the Adirondacks. In addition to being proximate to elevated emission sources, the Northeast is particularly sensitive to atmospheric deposition of Hg because of watershed and lacustrine characteristics (e.g., forest vegetation, shallow surficial deposits, wetlands, unproductive aquatic ecosystems) coupled with effects from acidic deposition.

Historical estimates of Hg emissions are uncertain but US anthropogenic emissions appear to have decreased approximately 38% since 1990, largely due to controls on medical waste incinerators and municipal waste combustors (www.epa.gov/ttn/atw/112nmerc/volume2.pdf). There are also plans to control Hg emissions from electric utilities within the next decade (www.epa.gov/mercuryrule/). In addition to reductions in atmospheric Hg deposition, fish Hg concentrations may be influenced by atmospheric deposition of strong acids. Across eastern North America, investigators have reported a negative correlation between fish Hg concentrations and lake pH from spatial surveys (Grieb et al. 1990; Spry and Wiener 1991; Driscoll et al. 1995). The Adirondack region of New York has been heavily affected by acidic deposition (Driscoll et al. 1991). However, recent controls on emissions of sulfur dioxide (SO_2) have resulted in reductions in precipitation and surface water sulfate (SO_4^{2-}), and some improvements in the acid–base status of some Adirondack lakes (Driscoll et al. 2003a, b).

There is keen interest in understanding the response of forested watersheds and lakes to changes in air emissions of Hg. Unfortunately, there is very limited information available to understand and quantify the critical linkages between changes in Hg emissions and response of ecosystems. In general spatial patterns have been used to infer how Hg might respond to decreases in atmospheric Hg deposition and changes in environmental conditions (e.g., changes in acidic deposition, changes in land use). There have been few long-term studies of trends in Hg concentration. Most studies tracking the recovery of Hg contaminated aquatic ecosystems have focused on systems with point sources (see Munthe et al. 2007 for a recent synthesis). There are only a few published reports of changes in fish Hg concentrations in response to changes in atmospheric Hg loading. Hrabik and Watras (2002) reported an ~30% decrease in yellow perch (*Perca flavescens*) Hg in a seepage lake (Little Rock Lake, WI, USA) in response to decreased atmospheric Hg deposition and in part due to decreased SO_4^{2-} deposition. Johansson et al. (2001) found northern pike (*Esox lucius*) Hg concentrations declined ~20% in 42 remote Swedish lakes, as wet Hg deposition declined by more than 50%. In another study of 176 Minnesota lakes (USA), there was a mixed result in the response of predatory fish Hg levels to decreased atmospheric Hg loading, with 44 lakes showing an increase, 87 lakes showing a decrease, and 45 lakes with no change in fish Hg concentrations (MPCA 2007). Drevnick et al. (2007) reported a decline in northern pike Hg levels in the lakes of Isle Royale (USA) in response to decreased SO_4^{2-} deposition.

In this study, we evaluated the response of yellow perch Hg concentrations in the Adirondacks to changes in atmospheric deposition of Hg and strong acids by resurveying the same cluster of 25 Adirondack lakes that were originally surveyed in 1992–1993 by Driscoll et al. (1994, 1995). We evaluated if spatial patterns of water column and fish Hg concentrations could serve as a surrogate for temporal trends. We hypothesized that Hg concentrations in lake water and yellow perch would have declined in the 13 years since the 1992–1993 survey, and that the greatest reductions in fish Hg would be those lakes with the largest decreases in water column MeHg and greatest increases in pH. The objectives of this study were: (1) to quantify changes in water chemistry and fish Hg concentration from 1992–1993 to 2005–2006; and (2) to investigate ancillary water chemistry parameters and watershed and lacustrine characteristics that influence changes in Hg species in the water column and fish Hg concentrations.

Study site and methods

The Adirondack Park is located in northern New York state and encompasses a large (~24,000 km^2) predominately forested portion of the Canadian Shield underlain by metasedimentary rocks and granite gneisses. The soils of the Adirondacks are largely Spodosols developed from glacial surficial materials that cover much of the region (Driscoll et al. 1994; Sullivan et al. 2005). The Park has a humid continental climate with short cool summers and long cold winters, and receives approximately 100 cm of precipitation annually with greater values at high elevations and toward the west (Driscoll et al. 1991). There are approximately 2,800 lakes in the Adirondack Park (Driscoll et al. 1991). Ice coverage of lakes usually persists from November to April.

A cluster of 25 lakes, located in the Adirondacks, were chosen by Driscoll et al. (1994, 1995) for a spatial survey of Hg concentrations in the yellow perch and the water columns. We resurveyed these same 25 lakes in 2005–2006 to investigate if changes in yellow perch and water column Hg concentrations have occurred. The yellow perch is a common indicator fish for Hg studies (Suns and Hitchin 1990; Rose et al. 1999; Greenfield et al. 2001) and is widely distributed throughout Adirondack lakes, as well as the upper Midwest and southeastern Canada. The yellow perch is also a popular commercial and recreational species and is widely sought for both human and wildlife consumption.

The 25 lakes for this study were chosen as a subset from 1469 Adirondack lakes that were originally surveyed by the Adirondack Lake Survey Corporation (ALSC) from 1984 to 1987 (www.adirondacklakessurvey.org). The ALSC collected physical, chemical, and biological data on each lake and its watershed and developed a classification system based on characteristics of hydrologic flow paths, surficial geology, and concentrations of organic solutes. The 25 lakes for this study were chosen to represent a broad range of the ALSC lake classes including those where high concentrations of Hg are thought to occur in fish tissue (e.g., seepage lakes, acidic lake classes such as thin till drainage lakes, high dissolved organic carbon (DOC) lakes). Two of the 25 lakes (Sunday Lake and Halfmoon Lake) exhibit complete or nearly complete depletion of O_2 in the hypolimnion during summer stratification (Driscoll et al. 1994). Lake and watershed characteristics for each of the 25 lakes in this study were taken from the Adirondack Lake Resurvey Corporation's website (www.adirondacklakessurvey.org). The original 1992–1993 survey and 2005–2006 resurvey were conducted from August to September.

Water sample collection and analysis

We used the same clean sampling protocols used by Driscoll et al. (1994, 1995) in the earlier survey to ensure comparability between collections. Water was collected as a grab sample over the side of a non-metallic boat at 0.25 m depth with 1 l Teflon bottles for Hg analysis and 1 l polypropylene bottles for ancillary chemistry analysis in accordance with US EPA sampling method 1669 (United States Environmental Protection Agency 1996) for trace metals. The bottles were opened and closed at 0.25 m depth and were rinsed three times with water prior to collecting the final sample. Each of the 25 lakes was sampled once during the original survey by Driscoll et al. (1994, 1995) and once again during the 2005–2006 resurvey.

Water samples were analyzed for total mercury (THg) utilizing automated oxidation, purge and trap, desorption, and cold-vapor atomic fluorescence spectrometry (CVAFS) (Tekran 2600; Tekran, Toronto, ON, USA) following US EPA method 1631, revision E (2002). Water samples were analyzed for MeHg utilizing distillation, aqueous ethylation, purge and trap, desorption, and CVAFS detector (Tekran 2500; Tekran, Toronto, ON, USA; Perkin Elmer Clarus 500 Gas Chromatograph; Perkin Elmer, Inc., Waltham, MA, USA) in accordance with US EPA method 1630 (2001). Lake water pH and acid neutralizing capacity (ANC) were determined in the laboratory using a Brinkmann Metrohm 716 DMS Titrino and 760 sample changer with a Ross General Purpose Sure-Flow pH electrode (Brinkmann Instruments, Mississauga, ON, USA). Lake water SO_4^{2-} samples were analyzed using ion chromatography (Dionex Corp, Sunnyvale, CA, USA) (Tabatabai and Dick 1983). Water samples were prepared for DOC analysis by filtering through a Whatman glass microfiber filter (934-AH, 1.5 µm pore size) and analyzed using infrared detection following persulfate oxidation for DOC (Dorhman Phoniex 8000 UV—Persulfate TOC Analyzer, Teledyne Tekmar, Mason, OH, USA) (Menzel and Vaccaro

1964). Lake water samples were analyzed for monomeric aluminum (Al_m) colorimetrically following chelation with pyrocatecholviolet (McAvoy et al. 1992). Non-labile (organic) monomeric aluminum (Al_o) was analyzed using the same method as Al_m, after passing the sample through an ion exchange column. Inorganic monomeric aluminum (Al_i) was calculated as the difference between Al_m and Al_o.

Quality assurance procedures for water chemistry analysis during both survey periods included field and sample duplicates, sample and trip blanks, continuous calibration verification (CCV), continuous calibration blank (CCB), matrix spike (MS), and matrix spike duplicates (MSD) within each batch of 20 samples. The accuracy and precision of the spikes and standards were maintained within the acceptable recoveries for the method guidelines (10% on CCV, MS, and MSD). Instrument performance and stability during analysis were evaluated with initial and ongoing precision recovery, instrument detection limit, and second source verification.

Yellow perch sampling and THg analysis

As in the earlier survey, yellow perch were collected by a combination of angling and experimental Swedish variable-mesh gill nets (length = 45.5 m; depth = 1.5 m) consisting of six panels (panel length = 7.5 m) with six mesh sizes (38, 51, 60, 64, 76, and 89 mm). Live, undamaged yellow perch were selected for analysis. Approximately 30 yellow perch from each of the 25 lakes were analyzed for Hg during the original survey (1992–1993; $n = 725$), while approximately 60 yellow perch from each lake were analyzed during the resurvey (2005–2006; $n = 1{,}155$). Total length (to 1 mm) and weight (to 0.01 g) measurements were obtained for each fish. The yellow perch were aged utilizing both scales and opercular bones. Fish were individually wrapped in aluminum foil, placed in Ziploc bags, and kept on ice until they could be frozen (within 4–8 h). The fish were frozen and stored at −20°C until analyzed.

Fish were removed from the freezer and allowed to partially thaw prior to tissue sampling. Tissue samples were collected from the dorsal musculature of each fish by first removing the scales and skin with a scalpel. The chemical form of Hg in fish is typically about 95% MeHg (Bloom 1992); therefore, fish tissue samples during both survey periods were analyzed for THg. During the 1992–1993 survey, Driscoll et al. (1994) removed ~1.0 g of tissue from each fish and analyzed digests of the fish tissue for THg utilizing automated oxidation, purge and trap, desorption, and (CVAFS). Sample duplicates, sample blanks, tuna fish mercury standards from the National Bureau of Standards, and matrix spikes were included within each batch of 20 samples to maintain quality assurance. For the 2005–2006 resurvey, a tissue plug of ~150 mg was removed from each fish using a disposable stainless steel 6 mm dermal punch (Biopunch, Fray Products, Buffalo, NY, USA). A complete description of fish tissue sampling procedures using a dermal punch is provided by Baker et al. (2004) and Peterson et al. (2005). During the 2005–2006 resurvey, fish tissue was analyzed for THg using a Milestone DMA80 (Direct Mercury Analyzer, Milestone, Monroe, CT, USA), in accordance with US EPA method 7473 (1998). A general overview of the instrument has been presented by Boylan and Kingston (1998). Two certified reference materials, DORM 2 (dogfish muscle tissue) and DOLT 2 (dogfish liver tissue), method and sample blanks, sample duplicates, and matrix spikes were run within each batch of 20 samples during the 2005–2006 resurvey.

Bioaccumulation factor (BAF) and fish body condition

A mean bioaccumulation factor (BAF) was calculated for each of the 25 lakes for each survey period. The BAF is expressed as the $\log_{10}$ ratio of yellow perch THg concentration to MeHg water column concentration. Relative weight (W_r) is an index of fish body condition and was calculated for each lake's population of yellow perch for each survey period. Relative weight values (W_r) are calculated by dividing the actual weight (W) of a fish by a standard weight (W_s) for a fish of that length and multiplying it by 100 ($W_r = (W/W_s) \times 100$; Willis and Guy 1991; Anderson and Neumann 1996). The length-specific standard weight (W_s) is predicted by a weight-length regression ($\log_{10} W_s = -5.386 + 3.230 \ \log_{10} L$) constructed by Anderson and Neumann (1996) specifically for yellow perch (W_s is weight (g) and L is total length (mm)). The mean W_r was used to interpret the overall nutritional/health status of the yellow perch population for a given lake during both the 1992–1993 and 2005–2006 surveys. A mean W_r of 100 for a population of yellow perch may reflect optimum ecological and physiological conditions for populations, a W_r below 100 may indicate problems in food or feeding conditions, and a W_r value above 100 may indicate that fish are not making the best use of a surplus of prey (Anderson and Neumann 1996).

Statistical analysis

All statistical procedures were performed using SAS Institute's software (Statistical Analysis Software, version 9.1, 2007, SAS Institute Inc., Cary, NC, USA). Our first objective was to gain a general understanding of changes in water chemistry and fish Hg concentrations for the 25 lakes from 1992–1993 to 2005–2006. For this analysis we compared the 1992–1993 water chemistry data (THg, MeHg, pH, ANC, SO_4^{2-}, DOC, Al) with the 2005–2006 water chemistry data. We calculated change for each water chemistry variable as the 1992–1993 concentration subtracted from the 2005–2006 concentration (e.g., $\Delta[\mathrm{THg}] = [\mathrm{THg}]_{05\text{–}06} - [\mathrm{THg}]_{92\text{–}93}$). In addition to comparing the individual lake concentrations over time, we also calculated and compared the median and range of water chemistry data across the 25 lakes in 1992–1993 to the 2005–2006 resurvey.

To investigate possible influences on water chemistry from years of extreme hydrologic conditions, we examined long-term (1960–2006) meteorological data for three National Climatic Data Center stations in the Adirondack Park (www.ncdc.noaa.gov/oa/ncdc.html). We used precipitation data as a surrogate for determining hydrologic conditions due to the absence of measured discharge and lake stage height data. We pooled data from the three stations to calculate mean annual precipitation and temperature values. A one way ANOVA was performed to determine if there were statistically significant differences ($P < 0.05$) among 1992, 1993, 2005, 2006 and long term (1960–2006) data.

We compared 1992–1993 yellow perch THg concentrations to 2005–2006 yellow perch THg concentrations. Our null hypothesis was that no difference existed between yellow perch THg concentrations for a particular lake between the original survey and the resurvey. A linear relationship exists between the THg concentration of yellow perch and fish length (Driscoll et al. 1994). Therefore, an analysis of covariance (ANCOVA) was performed using the SAS PROC GLM (generalized linear modeling) to test for differences for length-adjusted least squares mean yellow perch THg concentration between the original survey and the resurvey for each lake. A $\log_{10}$ transformation was used for yellow perch THg data to satisfy the assumption of normality and homoscedasticity. Wiener et al. (2006) suggested that younger yellow perch are better indicators of ecosystem processes and factors influencing the abundance of MeHg in lakes, rather than to substantial differences in trophic position. Therefore, a second similar statistical test was performed using only 1–2 year old yellow perch to investigate if younger age class yellow perch responded differently than the yellow perch population as a whole for a given lake. A separate analysis was conducted to compare fish THg concentrations across lakes by calculating a standard sized (20 cm) yellow perch THg concentration for each lake during both survey periods. We plotted for each individual lake and survey period the regression lines of Log_{10} fish Hg concentration against fish length. From each regression line we determined the predicted Hg concentration at the standard length (20 cm) for each lake during both survey periods.

The second objective of our statistical analysis was to investigate ancillary water chemistry parameters and watershed and lacustrine characteristics that influence Hg species in the water column and fish Hg concentrations. In the earlier survey, Driscoll et al. (1994, 1995) reported significant relationships between water column THg/MeHg and DOC and also strong relationships between yellow perch THg concentrations and lake water pH, Al_m, DOC, and % wetlands. We reevaluated the strengths of these relationships for the 2005–2006 resurvey.

Finally, we used multiple linear regression analysis (SAS PROC REG Stepwise) to construct models for predicting change in yellow perch THg concentration. Potential predictor variables included water chemistry (pH, ANC, SO_4^{2-}, DOC, Al, THg, MeHg), fish biology (BAF, fish body condition), and watershed and lacustrine characteristics (lake volume, mean lake depth, lake elevation, watershed area, lake surface area, WA:SA, % wetland area, % littoral area). A subset of potential predictor variables were chosen based on Pearson moment correlation coefficients. Addition and removal of variables into the model judged significant by the stepwise procedure was conducted based on P values ($P < 0.05$). The final selection of a

Table 1 Precipitation and temperature data for the Adirondack Park (1960–2006)

	Station name			
	Newcomb[a]	Tupper lake[b]	Lowville[c]	Mean
Annual precipitation (cm)				
1992	96.8	114.2	108.1	106.4
1993	95.6	114.1	100.0	103.3
2005	128.9	117.3	132.4	126.2
2006	116.8	126.3	115.2	119.4
Mean (1960–2006)[d]	104.4 (14.0)	102.6 (15.5)	102.9 (12.2)	103.3
Mean annual temperature (°C)				
1992	4.5	4.0	5.2	4.6
1993	5.1	4.6	5.8	5.2
2005	6.6	5.9	7.0	6.5
2006	6.5	6.3	7.7	6.8
Mean (1960–2006)	5.1	5.0	6.7	5.6

[a] Newcomb station, Essex County (43°58′N 74°11′W); elevation = 497 m

[b] Tupper lake station, Franklin County (44°14′N 74°26′W); elevation = 512 m

[c] Lowville station, Lewis County (43°48′N 75°29′W); elevation = 262 m

[d] Values in parentheses represent one standard deviation from the mean

multiple-linear regression model was based on several criteria including: model coefficient of determination (r^2), Adjusted r^2, and Akaike information criterion (AIC). Standardized residual plots of the final model were examined to satisfy assumptions of homoscedasticity and normality.

Results

Long term hydrologic data

Mean annual air temperatures in 2005 and 2006 exceeded the means for 1992, 1993 and the long-term record of 1960–2006 (Table 1). The 2005 precipitation was statistically greater ($P < 0.05$) than the 1992, 1993, 2006, and long term mean, with the exception of the Tupper Lake station (Table 1), indicating that 2005 was a relatively wet year. There was no statistical difference between the 1992, 1993, 2006, and long-term mean (1960–2006) precipitation amounts. While 2005 was a relatively wet year, the other 3 years of the survey (1992, 1993, 2006) were not statistically different from one another. It seems unlikely that hydrologic conditions between the original survey and resurvey varied enough to influence lake water and/or fish THg concentrations to an appreciable extent.

Changes in lake water chemistry from 1992–1993 to 2005–2006

The acid–base status of the 25 Adirondack lakes has changed markedly since the early 1990s. Lake water pH has increased in all but three lakes (Fig. 1a). The median pH for the cluster of 25 lakes has increased from pH = 6.26 in 1992–1993 (range = 4.94–7.09) to pH = 6.54 during the 2005–2006 resurvey (range = 5.02–7.36). Concurrent with increases in pH, the ANC has also increased in the 25 lakes. The median 1992–1993 ANC increased from 74.8 μeq l^{-1} in 1992–1993 to 91.0 μeq l^{-1} during the 2005–2006 resurvey. The range of ANC was large during both surveys (1992–1993 = −20.3–366.9 μeq l^{-1}; 2005–2006 = 5.2–319.7 μeq l^{-1}). The minimum ANC during both surveys was a thin-till drainage lake, Halfmoon Lake, and the maximum ANC was a salt-impacted lake, Tanaher Pond.

In addition to changes in pH and ANC, there were changes in other water chemistry parameters (e.g., SO_4^{2-}, DOC, Al) relevant to recovery status of aquatic ecosystems impacted by acidic deposition.

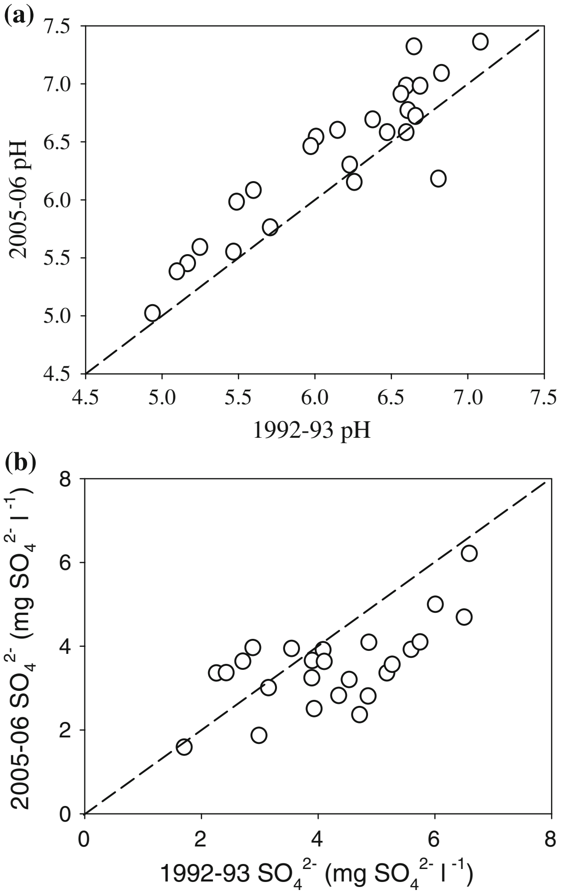

Fig. 1 **a** Lake pH in each of the 25 lakes in 2005–2006 compared with values in 1992–1993. **b** Lake sulfate (SO_4^{2-}) concentration (mg SO_4^{2-} l^{-1}) in each of the 25 lakes in 2005–2006 compared with values in 1992–1993. The *dashed line* represents a 1:1 line and each *circle* represents an individual lake in 1992–1993 and 2005–2006

Decreases in water column SO_4^{2-} concentration were measured in all but five of the resurveyed lakes (Fig. 1b). The median water column SO_4^{2-} concentration decreased from 4.1 mg SO_4^{2-} l^{-1} in 1992–1993 (range = 2.3–6.6 mg SO_4^{2-} l^{-1}) to 3.6 mg SO_4^{2-} l^{-1} during the 2005–2006 resurvey (range = 1.9–6.2 mg SO_4^{2-} l^{-1}). The DOC concentration decreased in 13 lakes, increased in 11 lakes, and showed no change in one lake. The median DOC across the 25 lakes decreased slightly from 7.0 mg C l^{-1} in 1992–1993 (range = 2.1–22.9 mg C l^{-1}) to 6.2 mg C l^{-1} in 2005–2006 (range = 2.7–19.0 mg C l^{-1}). The Al_m concentration decreased in all 25 lakes resurveyed. The median Al_m decreased markedly from 79.5 μg l^{-1} in 1992–1993 (range = 50.7–177.8 μg l^{-1}) to 28.2 μg l^{-1} (range = 14.3–121.9 μg l^{-1}) in 2005–2006. The Al_o fraction made up a much larger percentage (>80%) of Al_m than the Al_i fraction in the resurveyed lake water, coincidently there was also a large parallel decrease in the Al_o fraction in all 25 lakes. Because the Al_o was such a large fraction of Al_m compared to Al_i, most of the decrease in Al_m was measured in the Al_o fraction (~96% of the Al_m decrease). The Al_m concentration was negatively correlated with lake water pH (1992–1993 $r^2 = 0.59$ $P < 0.05$; 2005–2006 $r^2 = 0.67$ $P < 0.05$) and the Al_o fraction was positively correlated with lake water DOC concentration (1992–1993 $r^2 = 0.26$ $P < 0.05$; 2005–2006 $r^2 = 0.38$ $P < 0.05$).

Lake water THg decreased in 23 lakes (Fig. 2a) and MeHg concentrations decreased in 19 lakes (Fig. 2b). The median THg concentration decreased from 3.06 ng l^{-1} in 1992–1993 (range = 1.52–7.01 ng l^{-1}) to 1.82 ng l^{-1} in 2005–2006 (range = 0.56–5.07 ng l^{-1}). The median MeHg concentration decreased from 0.27 ng l^{-1} in 1992–1993 (range = 0.03–0.96 ng l^{-1}) to 0.14 ng l^{-1} in 2005–2006 (0.03–0.60 ng l^{-1}). The five lakes that had an increase in MeHg concentration did not have an increase in THg concentration, indicating for these sites an increase in the MeHg fraction occurred independent of a change in THg concentration. The MeHg fraction of THg in the water column ranged from 2 to 19% for both the original 1992–1993 survey and the 2005–2006 resurvey.

The water column DOC concentration has a significant role controlling both THg and MeHg concentrations as well as observed changes in Hg species. Both THg and MeHg water column concentrations were positively correlated with DOC concentration (Fig. 3). The slope of the regression line for THg–DOC and MeHg–DOC has decreased during 2005–2006 compared to the original 1992–1993 survey (Fig. 3), suggesting that there has been a decrease in Hg per unit DOC over the last 13 years. Change in lake water DOC concentration (calculated as the 1992–1993 concentration subtracted from the 2005–2006 concentration) was positively correlated to change in THg concentration ($r^2 = 0.60$ $P < 0.05$) and change in MeHg concentration ($r^2 = 0.18$ $P < 0.05$).

Yellow perch THg concentration and body condition

During the 1992–1993 survey, 64% of the total number of yellow perch analyzed ($n = 725$) exceeded the

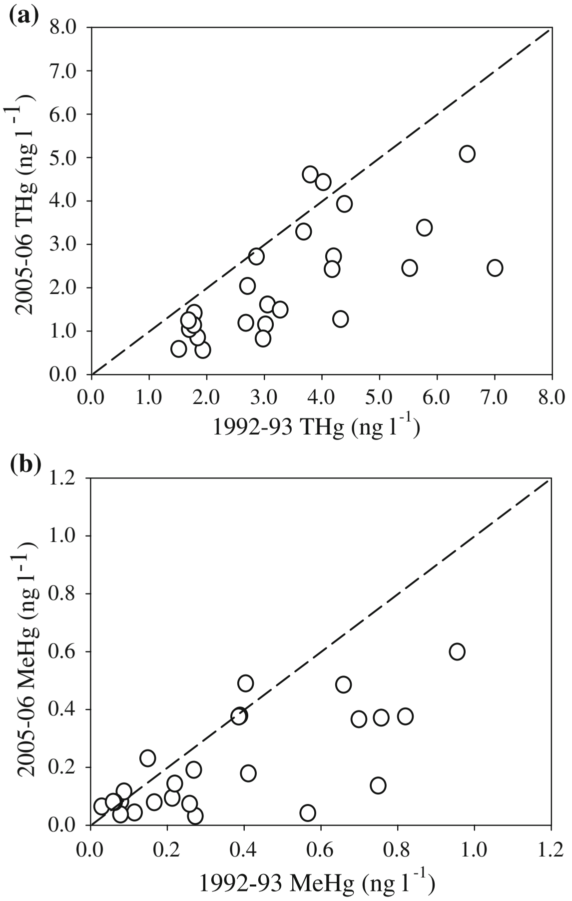

Fig. 2 **a** Lake water column total mercury (THg) concentration (ng l^{-1}) in 2005–2006 compared to 1992–1993. **b** Lake water column methyl mercury (MeHg) concentration (ng l^{-1}) in 2005–2006 compared to 1992–1993. The *dashed line* represents a 1:1 line and each of the *circles* represents an individual lake's Hg concentration in 1992–1993 and 2005–2006

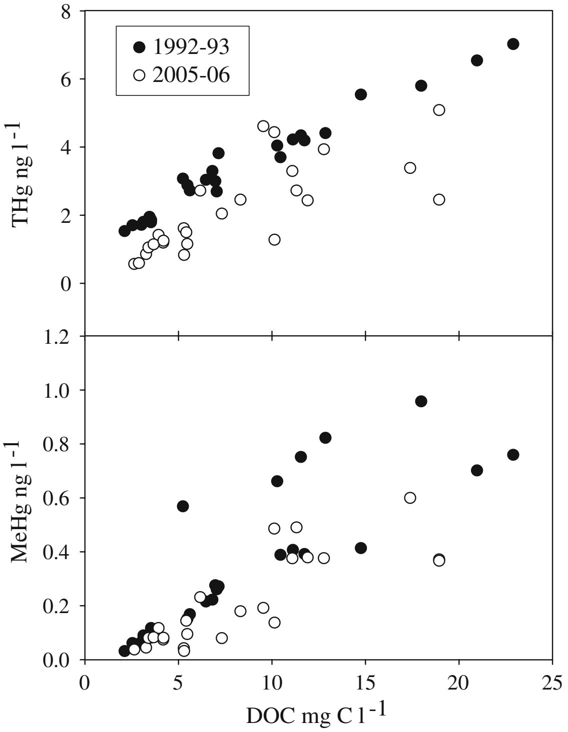

Fig. 3 Lake water total mercury (THg) and methyl mercury (MeHg) concentration (ng l^{-1}) as a function of dissolved organic carbon (DOC) concentration (mg C l^{-1}) for the 1992–1993 survey ($THg = 0.26DOC + 1.16$ $r^2 = 0.97$ $P < 0.05$; $MeHg = 0.04DOC + 0.0007$ $r^2 = 0.71$ $P < 0.05$) and the 2005–2006 resurvey ($THg = 0.20DOC + 0.50$ $r^2 = 0.57$ $P < 0.05$; $MeHg = 0.03DOC - 0.036$ $r^2 = 0.68$ $P < 0.05$)

US EPA tissue residue criterion for MeHg in fish tissue of 0.3 μg g^{-1} (wet weight) (US EPA 2006) assuming that all of the measured THg occurred as MeHg (Bloom 1992). In comparison, during the resurvey (2005–2006), 49% of the total number of yellow perch surveyed ($n = 1154$) exceeded the 0.3 μg g^{-1} THg concentration. The percentage of yellow perch among all 25 lakes that exceeded the US EPA tissue residue criterion for MeHg in fish tissue increased with fish age (Fig. 4a). Both surveys were designed to sample a complete range of age classes for yellow perch (i.e., 1–10 year old fish). Yellow perch age range and mean lengths were similar in each of the 25 lakes for the original 1992–1993 survey and 2005–2006 resurvey (Table 2). Yellow perch THg concentrations increased with fish age. The mean THg concentration among all yellow perch in all 25 lakes exceeded the 0.3 μg g^{-1} action limit by age two (Fig. 4b).

During the original 1992–1993 survey, mean yellow perch W_r values for the 25 lakes ranged from 68 to 86 (median = 76), compared to the 2005–2006 resurvey, W_r ranged from 71 to 99 (median = 78). Mean W_r in yellow perch increased in eight of the 25 lakes, decreased in four lakes, and did not change in four lakes (*t*-test; $P < 0.05$).

Factors influencing yellow perch THg concentration

Among the 25 lakes, mean yellow perch THg concentration was negatively correlated to lake pH (Fig. 5) and positively correlated with water column Al_m concentration (Fig. 6). The Al_o fraction of Al_m had a stronger correlation with yellow perch THg (1992–1993 $r^2 = 0.40$ $P < 0.05$; 2005–2006 $r^2 = 0.60$ $P < 0.05$)

Table 2 Number of yellow perch sampled, mean fish length, range of fish age and standardized 20 cm yellow perch total mercury (THg) concentration, and change in yellow perch THg concentration

Lake name	*n*		Length (cm)[a]		Age		THg ($\mu g\ g^{-1}$)[b]		Change[c]
	1992–1993	2005–2006	1992–1993	2005–2006	1992–1993	2005–2006	1992–1993	2005–2006	
North Pink	30	58	15.6 (1.2)	15.8 (1.8)	3–7	1–7	0.39	0.30	D
Middle	28	59	17.2 (6.0)	16.8 (2.9)	1–11	2–6	0.31	0.24	D
Little Weller	29	59	17.3 (2.3)	18.8 (3.4)	2–7	2–9	0.40	0.23	D
Tamarack	30	55	19.7 (4.9)	16.3 (3.9)	2–7	1–9	0.43	0.29	D
North	30	45	15.6 (4.5)	16.4 (1.9)	1–8	2–6	0.32	0.31	D
Halfmoon	30	45	18.2 (3.8)	17.6 (3.7)	2–12	2–8	0.69	0.36	D
Santa Clara	30	8	20.7 (5.8)	27.7 (3.7)	3–11	5–8	0.21	0.09	D
Rainbow	30	57	17.7 (4.0)	18.8 (3.6)	1–11	2–9	0.37	0.21	D
Haymeadow	28	39	21.3 (8.2)	22.1 (7.9)	2–8	1–7	0.92	0.48	D
Francis	30	71	19.8 (4.8)	16.4 (3.2)	2–12	1–5	0.55	0.39	D
Little Safford	27	22	15.5 (5.1)	20.7 (3.4)	1–7	1–5	0.49	0.32	D
Limekiln	28	67	15.5 (2.5)	17.1 (3.3)	1–6	1–4	0.53	0.34	D
Kushaqua	28	56	15.9 (4.3)	18.8 (3.8)	1–10	2–6	0.36	0.38	I
Fall	30	31	22.2 (3.3)	21.2 (3.9)	3–9	2–8	0.39	0.44	I
Oregon	28	51	25.9 (2.3)	25.3 (3.1)	2–10	2–10	0.09	0.13	I
Sand	28	60	17.1 (3.9)	15.5 (1.8)	1–6	1–7	0.51	0.75	I
Sagamore	28	25	14.6 (5.4)	16.5 (1.8)	1–10	2–5	0.59	0.67	I
Upper Sister	30	60	16.7 (2.3)	15.4 (1.4)	2–7	2–6	0.68	1.11	I
Tanaher	30	36	17.3 (1.4)	17.9 (4.6)	3–7	2–8	0.38	0.36	NC
Big Moose	27	44	16.0 (4.4)	18.4 (4.1)	1–8	2–8	0.79	0.74	NC
Hitchens	30	25	21.2 (5.7)	19.4 (4.7)	2–13	2–9	0.40	0.44	NC
Rondaxe	30	21	20.9 (6.3)	20.0 (4.2)	1–9	2–8	0.35	0.43	NC
Rock	30	71	17.7 (5.5)	21.5 (5.9)	1–12	1–8	0.40	0.45	NC
Sunday	29	53	19.2 (2.6)	17.0 (2.2)	1–10	1–4	0.96	1.08	NC
Moss	27	39	17.3 (5.3)	18.9 (4.7)	1–5	1–4	0.42	0.39	NC

[a] Data in parentheses represent one standard deviation from the mean

[b] Standardized 20 cm yellow perch THg concentration (wet/weight) predicted from the linear regression of Log_{10} transformed fish THg concentration against fish length

[c] Change was calculated using ANCOVA to test length adjusted least squares mean yellow perch THg concentration between the two survey periods (see "Methods"). *D* Decrease in fish THg; *I* increase in fish THg; *NC* no change in fish THg

compared to the Al_i fraction of Al_m (1992–1993 $r^2 = 0.24$ $P < 0.05$; 2005–2006 $r^2 = 0.13$ $P > 0.05$). Yellow perch THg concentration was positively correlated with DOC concentration up to 8 mg C l^{-1} ($r^2 = 0.28$ $P < 0.05$) during the 1992–1993 survey and up to 5 mg C l^{-1} ($r^2 = 0.65$ $P < 0.05$) during the 2005–2006 resurvey. Lake water THg water column concentrations were positively correlated during the 2005–2006 resurvey with yellow perch THg concentrations ($r^2 = 0.28$ $P < 0.05$), but not during the original survey. Water column MeHg concentrations were positively correlated with mean fish THg concentrations (1992–1993 $r^2 = 0.23$ $P < 0.05$; 2005–2006 $r^2 = 0.24$ $P < 0.05$).

Watershed and lacustrine characteristics (lake volume, mean lake depth, lake elevation, watershed area, lake surface area, WA:SA, % wetland area, % littoral area) were compared to yellow perch THg concentrations. For both survey periods, the percent wetlands of the entire watershed and the percent wetland area of the near-shore area, was the strongest predictor of yellow perch THg concentration. The remaining watershed and lacustrine characteristics were not significantly correlated to fish Hg concentration for either survey period.

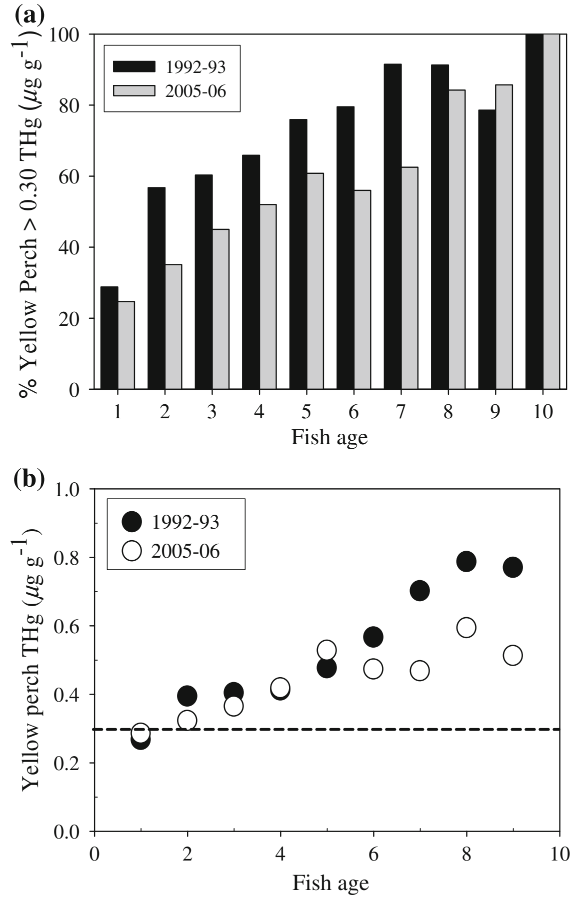

Fig. 4 **a** Percentage of yellow perch by age class for all 25 lakes that exceed the 0.3 μg g^{-1} US EPA tissue residue criteria for methyl mercury for 1992–1993 and 2005–2006. **b** Yellow perch mean total mercury (THg) concentration (μg g^{-1} wet weight) by fish age class for all 25 lakes (*dashed line* represents the 0.3 μg g^{-1} US EPA tissue residue criteria for methyl mercury) for 1992–1993 and 2005–2006

Among lakes with wetland area greater than 10% of the entire watershed ($n = 7$) THg in yellow perch was positively correlated to percent wetland (1992–1993 $r^2 = 0.56$ $P < 0.05$; 2005–2006 $r^2 = 0.28$ $P < 0.05$). Among lakes with a wetland area greater than 15% of the near shore area ($n = 10$), THg in yellow perch was positively correlated to percent wetland (1992–1993 $r^2 = 0.33$ $P < 0.05$; 2005–2006 $r^2 = 0.55$ $P < 0.05$).

Bioaccumulation of mercury in fish

The BAF between THg in yellow perch and MeHg in lake water was greater (*t*-test; $P < 0.05$) during the 2005–2006 resurvey in 14 of the lakes, decreased in four of the lakes, and did not change in seven of lakes.

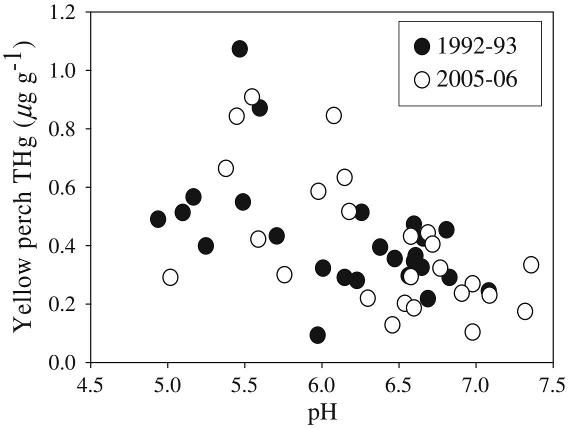

Fig. 5 Mean fish total mercury (THg) concentrations (μg g^{-1} wet weight) as a function of lake pH (1992–1993 $r^2 = 0.25$ $P < 0.05$; 2005–2006 $r^2 = 0.34$ $P < 0.05$)

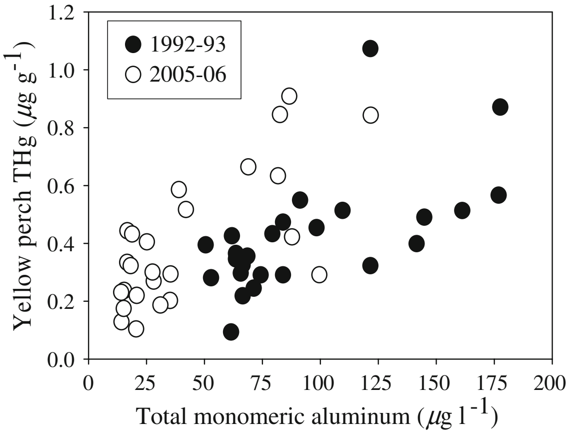

Fig. 6 Mean fish total mercury (THg) concentrations (μg g^{-1} wet weight) as a function of total monomeric aluminum concentration in lake water (μg l^{-1}) for each of the 25 lakes (1992–1993 $r^2 = 0.38$ $P < 0.05$; 2005–2006 $r^2 = 0.52$ $P < 0.05$)

The $\log_{10}$BAF for the 1992–1993 survey ranged from 5.66 to 6.86 (median BAF = 6.12) in the 25 lakes, compared to the 2005–2006 resurvey, the $\log_{10}$BAF ranged from 5.63 to 6.85 (median BAF = 6.39). The BAF was negatively correlated with DOC water column concentration (1992–1993 $r^2 = 0.61$ $P < 0.05$; 2005–2006 $r^2 = 0.61$ $P < 0.05$) and % MeHg of THg in the water column (1992–1993 $r^2 = 0.46$ $P < 0.05$; 2005–2006 $r^2 = 0.68$ $P < 0.05$).

Change in yellow perch THg

The change in yellow perch THg concentrations varied among lakes. Yellow perch THg concentrations decreased in 12 lakes ($P < 0.05$), increased in six lakes ($P < 0.05$), and did not change in seven lakes ($P > 0.05$). A second statistical testing, using only 1–2 year old yellow perch matched the results from the above analysis that included all age class yellow perch, with the exception of one lake, Limekiln Lake. In Limekiln Lake, the 1–2 year old yellow perch THg decreased, while including all age classes of yellow perch resulted in no significant change ($P > 0.05$) in fish Hg concentrations. The results from the two tests indicate that THg concentrations in the younger age classes of yellow perch responded in a similar manner as all age classes of yellow perch.

Multiple-linear regression analysis

The one variable in particular that was most effective in predicting change in yellow perch THg across the 25 lakes was fish condition. Change in fish THg concentration was positively correlated to change fish condition ($r^2 = 0.42$ $P < 0.05$), so that fish with lower THg concentrations in 2005–2006 compared to 1992–1993 were fish with increases in body condition. Lake elevation was also a significant variable for predicting change in fish Hg. Change in fish THg concentration was negatively correlated ($r^2 = 0.32$ $P < 0.05$) to lake elevation, so that those fish with an higher THg concentrations in 2005–2006 compared to 1992–1993 were in greater elevation lakes. We were better able to predict change in yellow perch (ΔYP) THg using a stepwise multiple linear regression model that include four significant variables (Table 3): watershed area (WA), lake elevation (ELEV), change in H^+ concentration ($\Delta[H^+]$), and change in yellow perch relative weight (ΔW_r).

$\Delta YP\ \ THg = -0.368 + 0.000012\ \ WA + 0.000608\ \ ELEV - 41085\ \ \Delta[H^+] - 0.009\ \ \Delta W_r$ ($r^2 = 0.69$ $P < 0.05$; $n = 25$ lakes).

This multiple regression model predicts that yellow perch THg concentration increases with greater lake watershed area and higher elevation, and decreases in lake pH and decreases in yellow perch body condition.

Table 3 Statistics for multiple linear regression equation

Predictor variable	β	SE[a]	P
Watershed area (km^2)	0.0012	0.0003	0.002
Elevation (m)	0.00061	0.00024	0.02
$\Delta[H^+]$ (μmol l^{-1})	−41,085	14,419	0.04
ΔW_r	−0.009	0.004	0.01

[a] *SE* is the standard error for the regression slope, β

Discussion

The results from this study represent one of the few published reports tracking changes in fish Hg, particularly large synoptic surveys of remote areas such as the Adirondacks. The significant increases in pH and ANC and decreases in Al_m concentrations in the 25 lakes surveyed are consistent with other reports of the chemical recovery of Adirondack lakes (Driscoll et al. 2007b) and lakes across the northeastern US (Warby et al. 2005). Concurrent with changes in the acid–base status of these lakes, there have also been decreases in THg and MeHg in the water column. Decreased water column THg may be partially attributed to recent decreases in U.S. Hg emissions due to controls on medical waste incinerators and municipal waste combustors (Driscoll et al. 2007a). Lorey and Driscoll (1999) observed declines in Hg deposition in Adirondack lake sediments over the last few decades. The decline in MeHg concentration is most likely in part due to decreased THg loading to the lakes but also due to watershed processes (e.g., hydrology, water chemistry, percentage wetlands, SO_4^{2-} inputs) that influence methylation/demethylation rates and the fluvial transport of both THg and MeHg.

We hypothesized that decreases in water column THg and MeHg would result in decreases in fish Hg in Adirondack lakes. However, despite the fact that these lakes are in close proximity to one another and receive approximately the same atmospheric Hg deposition, the changes in Hg concentrations of individual lakes was varied. There are multiple factors including atmospheric Hg deposition that could affect MeHg production, thus confounding the interpretation of the response of fish Hg to decreased atmospheric Hg deposition (Wiener et al. 2007). To better understand the response of aquatic ecosystems to decreases in atmospheric Hg loading we examined factors such as hydrologic conditions, lake acid–base status, changes in SO_4^{2-} and DOC water column concentrations that have been reported as important controllers of fish Hg (Gilmour and Henry 1991; Driscoll et al. 1995; Wiener et al. 2006).

Water chemistry response

Hydrologic conditions to a large degree regulate the acid–base chemistry and Hg concentration of surface waters with greater Hg concentrations during wetter years (Kamman et al. 2004; Evers et al. 2007). There were slightly warmer and wetter conditions during 2005, which would promote greater watershed to lake transport of solutes in runoff and enhance methylation in warmer sediments and soils. However, water column THg and MeHg concentrations were lower in 2005–2006 than the 1992–1993 survey suggesting that other factors besides a slightly wetter year in 2005 explain the spatial and temporal patterns. Moreover Selvendiran et al. (2008) data showed little variability in the THg and MeHg drainage water concentration and flux (August 2004 to July 2006) at the inlet and outlet of two Adirondack lakes located among our cluster of 25 lakes. Lawrence et al. (2004) also reported that there was no trend in daily mean flows in the Adirondack Region of New York for the 1990s.

Sulfate reducing bacteria (SRB) are important mediators of Hg methylation rates in aquatic ecosystems and therefore changes in SO_4^{2-} may alter net methylation rates and MeHg supply (Gilmour and Henry 1991; Benoit et al. 2003). There is evidence that SO_4^{2-} stimulates net Hg methylation in lacustrine sediments (Gilmour and Henry 1991). Results from SO_4^{2-} amendments to soil (Gilmour et al. 1992), and lakes and wetlands (Branfireun et al. 1999; Hrabik and Watras 2002; Jeremiason et al. 2006) have also reported increases in MeHg production. Drevnick et al. (2007) suggested that reductions in SO_4^{2-} deposition alone could explain decreases in fish Hg from Isle Royal lakes. A significant decrease in wet SO_4^{2-} deposition to the Adirondacks has occurred over the last two decades in response to reduced SO_2 emissions as mandated by the 1970 and 1990 Amendments of the Clean Air Act (CAAA) (Warby et al. 2005; Driscoll et al. 2007b). Due to the intimate relationship between Hg and the sulfur cycle, we initially hypothesized that the measured decrease in lake water SO_4^{2-} concentrations would lead to lower MeHg concentrations and decreased fish Hg. However, the change in water column SO_4^{2-} concentration was not correlated to change in water column MeHg concentration nor fish Hg concentrations.

Despite a strong correlation between both water column THg–DOC and MeHg–DOC (Fig. 3), the relationship between DOC and fish Hg was inconsistent. In the resurvey, yellow perch THg concentration was positively correlated with DOC up to 5 mg C l^{-1}. This pattern is consistent with Driscoll et al. (1995) who reported a positive correlation of fish THg with DOC up to 8 mg C l^{-1}. At DOC concentrations greater than 5–8 mg C l^{-1}, the relationship between fish Hg and DOC was weaker ($r^2 = 0.26$) suggesting that higher levels of DOC alter the availability of Hg. This is likely a result of the complex role DOC plays in Hg biogeochemistry.

It is well established that Hg has a strong affinity for organic matter with much of the binding of Hg to natural organic matter largely associated with reduced S groups (Haitzer et al. 2002; Skyllberg et al. 2003). The binding of Hg by DOC simultaneously facilitates the transport of Hg, and controls the availability of Hg for methylation and bioavailability to fish and other aquatic organisms (Driscoll et al. 1994, 1995; Hudson et al. 1994). The decrease in Hg per unit DOC (Fig. 3) is consistent with a regional long-term decrease in Hg deposition. In northern hardwood forests, the forested canopy is a major receptor of Hg due to the large surface area of the canopy (Rea et al. 2002; Bushey et al. 2008). Mercury is subsequently deposited to the forest floor via litterfall, which can constitute the most significant flux of Hg to the forest floor (St. Louis et al. 2001; Rea et al. 2002; Sheehan et al. 2006). It seems likely that decreases in atmospheric Hg deposition would be manifested through decreases in fluvial Hg transport by DOC.

Long-term changes in DOC were inconsistent across the lakes surveyed, with about half of the lakes with increases in DOC concentrations and about half of the lakes with decreases in DOC concentrations. In contrast the routine monitoring of Adirondack lake chemistry shows generally increases in DOC coincident with decreases in lake SO_4^{2-} (Driscoll et al. 2007b). While the mechanisms responsible for change in lake water DOC are not clear, researchers have suggested it may be linked to decreases in acidic deposition (Monteith et al. 2007). Regardless of the mechanism, it is likely that any change in DOC will affect water column THg and MeHg concentrations and possibly the bioaccumulation of Hg in aquatic biota as well. The bioavailability of MeHg appears to be primarily regulated by the extent that Hg binds with organic acids (Driscoll et al. 1995). For example, the BAF was negatively correlated to DOC concentration which we hypothesize is due to greater amounts of DOC binding MeHg in the water column, thus limiting MeHg for uptake by aquatic organisms. The relationship between BAF and DOC complicates the linkage between changes in atmospheric Hg deposition and changes in fish THg concentrations.

While the relationship between DOC and fish THg is inconsistent, Adams et al. (2009) found a positive relationship between DOC and zooplankton Hg ($r^2 = 0.48$ $P < 0.05$) and zooplankton Hg and water column THg ($r^2 = 0.58$ $P < 0.05$) and MeHg ($r^2 = 0.42$ $P < 0.05$) in a 2005 survey of nine of the same lakes that studied here. For these nine lakes, zooplankton Hg was also correlated to yellow perch THg ($r^2 = 0.32$ $P < 0.05$). This pattern is consistent with Hg transport to lakes via DOC and then assimilated by zooplankton where Hg is then bioaccumulated to fish. Mercury concentrations in smaller size classes of seston in these nine lakes were also positively correlated to DOC (0.2–2.0 µm $r^2 = 0.33$ $P < 0.05$; 2.0–20.0 µm $r^2 = 0.36$ $P < 0.05$) and water column THg (0.2–2.0 µm $r^2 = 0.94$ $P < 0.05$; 2.0–20.0 µm $r^2 = 0.55$ $P < 0.05$) (Adams et al. 2009). This result emphasizes the important role of DOC regulating Hg transport and bioavailability particularly for aquatic organisms that consume zooplankton such as young yellow perch. Yellow perch smaller than 10 cm feed directly on plankton and littoral invertebrates, while yellow perch between 10 and 20 cm begin to consume other fish and invertebrates (Driscoll et al. 1994; Chen et al. 2005). The feeding preference of young yellow perch and older yellow perch is reflected in the fact that THg concentrations increase with length, weight, and age (Fig. 4). This pattern reflects change in prey selection and bioaccumulation of THg along the aquatic food chain for yellow perch (Driscoll et al. 1994).

Acidic deposition mobilizes elevated concentrations of Al_i from the soil to drainage waters in acid-sensitive areas like the Adirondacks (Cronan and Schofield 1990; Driscoll et al. 2003b). Increases in lake water pH and lower Al concentrations are associated with increased biodiversity and abundance of plankton, invertebrates, and fish in acid-sensitive waters (Driscoll et al. 2001). We observed a positive correlation between fish THg and water column Al concentration despite the fact that Al concentrations were lower in all 25 lakes during the resurvey. Simonin et al. (2008) recently reported a similar relationship for fish THg and Al in the Adirondack and Catskill mountains. This relationship may imply that Al is affecting MeHg binding on DOC. Metals such as Al complex with DOC and may alter the binding of Hg with organic ligands thus leading to greater bioavailability of MeHg in lakes with high Al:DOC ratios (Driscoll et al. 1994). While Al:DOC ratios have decreased substantially in all 25 lakes, it does not appear that DOC is now transporting greater quantities of Hg per unit of DOC as both THg:DOC and MeHg:DOC ratios have also decreased in all 25 lakes. An alternative hypothesis is that elevated Al and low pH associated with acidic deposition stresses fish and increases in pH and decreases in Al have allowed for increases in fish condition (see below).

Yellow perch response

The 25 Adirondack lakes in our study exhibited a mixed response to decreases in atmospheric deposition of Hg and strong acids. A significant decrease in yellow perch THg was measured in 12 of the 25 lakes, however, 18 of these lakes as recent of the 2005–2006 resurvey still have yellow perch THg concentrations (standardized for a 20 cm yellow perch) at or exceeding the 0.3 µg g^{-1} US EPA tissue residue criterion for MeHg for fish tissue (Table 2). In addition to the adverse human health impacts of high fish Hg concentrations, there are also potential negative health implications for wildlife. Young yellow perch are a preferred prey of common loons (*Gavia immer*) which are obligatory piscivores. The relative high percentage of young yellow perch (25% of 1 year old fish and 35% of 2 year old fish) that exceed the 0.3 µg g^{-1} criterion during the 2005–2006 resurvey (Fig. 4a) could adversely affect the reproductive success of loons nesting on these lakes. Burgess and Meyer (2008) reported that production of young common loons declined 50% when mercury levels in prey fish were 0.21 µg g^{-1} and that reproduction failed completely when mercury concentrations in prey fish were 0.41 µg g^{-1}.

Elevated concentrations of fish THg in Adirondack lakes are consistent with recent surveys that have identified the western and central Adirondacks as a biological Hg “hotspot” (Evers et al. 2007). The Adirondacks have among the highest concentrations of fish Hg in New York state (Simonin et al. 2008) and northeastern North America (Kamman et al. 2005). Adirondack lakes are susceptible to high concentrations of Hg in fish due to high Hg deposition (Miller et al. 2005) and elevated acidic deposition (Ito et al. 2002). Watershed characteristics such as an abundance of forest and wetland cover and oligotrophic and acidic conditions in Adirondack lakes likely lead to higher levels of Hg bioaccumulation in fish and wildlife in the Adirondacks (Driscoll et al. 2007a; Evers et al. 2007).

Fish THg concentrations can vary substantially between nearby lakes in the Adirondacks (Simonin et al. 2008) and it appears from our study that the response of yellow perch THg to changes in Hg and strong acid deposition can also vary among lakes within close proximity to one another. Therefore, variation in individual lake-watershed characteristics may regulate the change in fish Hg. Our multiple-linear regression model resulted in four significant lake-watershed characteristics for predicting change in yellow perch THg levels (lake elevation, watershed size, change in lake water pH, and change in the relative weight (W_r) of fish—an index of fish body condition).

Higher elevation areas in the Adirondack Park generally receive greater precipitation and SO_4^{2-} deposition that exposes lakes at higher elevations to greater acidic deposition (Ito et al. 2002). Similarly, atmospheric Hg deposition also increases with elevation for the Northeast (Miller et al. 2005). Higher elevation areas generally have thinner base-poor soil and have less productive lakes which are attributes conducive to high concentrations of Hg in the water column and fish.

Watershed characteristics to a large degree control the retention of atmospherically deposited Hg and the subsequent transport of Hg to downstream sites of methylation (i.e., wetlands, lakes) (Driscoll et al. 2007a). Watershed Hg retention is predicted be to greater in larger watershed areas, so that as watershed size increases yields of Hg decrease (Grigal 2002). Similarly, the watershed area to surface-water area ratios (WA:SA) have also been reported as important determinants of Hg retention and delivery to aquatic ecosystems (Munthe et al. 2007). Lakes with a small watershed relative to lake surface area are predicted to be more responsive to changes in atmospheric Hg deposition. Results from a recent watershed Hg isotope addition experiment demonstrate that fish Hg concentrations can respond rapidly to direct addition of Hg to the lake surface, however, a full response to atmospheric Hg deposition is delayed by the gradual export of Hg stored in watersheds (Harris et al. 2007). Terrestrial ecosystems may act as a time lag for the transport of atmospheric Hg deposition to lakes further confounding the relationship between Hg deposition and fish Hg (Munthe et al. 2007).

The results from our survey are in agreement that watershed area is an important variable in aquatic ecosystem Hg studies. However, we have observed that increases in fish Hg concentrations were correlated with increasing watershed area, while, no relationship exists between change in fish THg levels with WA:SA ratios. This finding is counterintuitive to the hypothesis that large watersheds are less responsive to changes in atmospherically deposited Hg. There is a general lack of understanding into the dynamics of Hg release from soil (Munthe et al. 2007) making quantitative predictions of the response of Hg concentrations in aquatic ecosystems uncertain. Larger watersheds may have larger zones for methylation (i.e., anoxic zones including wetlands and hydric soils) that may better connect hydrologically through flowpaths to the lake, thereby supplying greater amounts of MeHg than smaller watersheds. Both the amount of Hg stored in soil and the subsequent transport to downstream lakes as well as the hydrologic connectivity of watersheds are highly influential factors dictating the timing and magnitude of future change in Hg levels in aquatic ecosystems.

The strong correlation between fish Hg and lake water pH is agreement with other studies that have found higher fish Hg concentrations in lower pH lakes (Suns and Hitchin 1990; Driscoll et al. 1994). The methylation of ionic Hg is pH-dependent with a maximum production around pH 6; therefore, more production of MeHg occurs in lower pH waters (Suns and Hitchin 1990). Driscoll et al. (1994) proposed several linkages between surface water acidification and fish Hg concentration that may help explain the importance of water column pH as a significant variable in fish Hg studies. First, there may be increased partitioning of Hg to the water column in acidic water, so that as lake pH decreases there is more Hg available for methylation and uptake by aquatic organisms. Secondly, as the acidity in lakes is attenuated, the lakes may become more productive providing greater biomass and water quality conditions less stressful to fish leading to better fish condition. There also may be less production of MeHg and/or increased rates of demethylation and volatilization of Hg^o from the water column with increases in the pH of Adirondack lakes.

Fish condition is an important index of the Hg status of fish, with a better fish body condition having a lower Hg concentration (Suns and Hitchin 1990; Greenfield et al. 2001). Lakes with lower a pH (i.e., more acidic lakes) may have fish with a lower body condition due to reduced availability of preferred food items (Fechhelm et al. 1995; Greenfield et al. 2001). Earlier reports have demonstrated a negative relationship between fish Hg and body condition (Suns and Hitchin 1990; Greenfield et al. 2001). Fish body condition, while correlated to fish growth rates, is actually a better index of current environmental conditions than growth rates because body condition can change rapidly in response to environmental perturbation (Mills et al. 2000; Greenfield et al. 2001). Fish with a better body condition have reached a given size more efficiently (i.e., having spent less energy consuming and respiring to reach that size) and therefore have lower tissue contamination per body mass (de Freitas et al. 1974; Norstrom et al. 1976; Greenfield et al. 2001). As a consequence fish growth rates may not necessarily change in response to acidification (Hrabik and Watras 2002), but the fish body condition may be lower. Therefore, faster growing fish can exhibit "growth dilution" of tissue contaminates such as Hg (Greenfield et al. 2001).

Conclusions

Yellow perch THg concentrations have decreased in 12 of the resurveyed lakes, increased in six lakes, and did not change significantly in seven lakes. However, the mechanism(s) responsible for changes in fish THg is not entirely clear. In contrast to our initial hypothesis, decreased water column MeHg concentration was not directly correlated to change in yellow perch THg concentrations. The four variables that were correlated to change in yellow perch THg were watershed area, elevation, change in pH, and change in fish body condition. As the acidity in lakes is attenuated in response to decreases in acidic deposition, lakes may become more productive and water quality conditions less stressful to fish. Improvement in the acid–base status of Adirondack lakes may lead to an increase in fish condition. As fish body condition increases, fish can exhibit "growth dilution" of tissue contaminants leading to lower fish THg concentrations. This pattern is particularly true for lower elevation lakes that receive lower amounts of acidic and Hg deposition than higher elevation lakes. The roles of watershed area and/or WA–SA ratios are not entirely clear. The response of ecosystems will likely be a mixture of responding to decreased Hg emissions, but the degree of this change may be limited by legacy Hg that is retained and subsequently transported from the watersheds.

Spatial and temporal studies indicate there remains a poor understanding of controls on the variability of fish Hg. From our study it appears that additional controls on Hg and acidic emissions will be necessary to accelerate the recovery of Adirondack lakes; a relatively large percentage (49%) of the yellow perch sampled have THg concentrations above 0.3 $\mu g\ g^{-1}$. There are several variables that appear important predictors for the response of fish Hg, however, it is unclear how much reduction in Hg and/or acidic deposition will be necessary to attain a full reduction in Adirondack fish tissue Hg levels to below the US EPA action level of 0.3 $\mu g\ g^{-1}$. Additional monitoring of drainage waters and lakes in the Adirondacks will be necessary to quantify the future response of aquatic ecosystems. A future survey of the 25 lakes in this study would allow for a more complete trend analysis to provide data on the continued response of yellow perch to changes in atmospheric deposition of Hg and strong acids.

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