

Trends in Mercury Concentrations in New York State Fish

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Abstract Atmospheric emissions of mercury in the US are being reduced, but worldwide mercury emissions remain high. Mercury is also being removed from many consumer items. Changes over time in mercury concentrations in fish remain important to resource managers and the general public. There is hope that mercury concentrations in fish will decline, and the number of fish consumption advisories due to mercury will decrease. We compared mercury concentrations in yellow perch (*Perca flavescens*) from a group of Adirondack lakes with data collected 12–17 years earlier and found variable responses among lakes. We observed an average decline of 14% in yellow perch mercury concentrations over the past 15 years.

Keywords Mercury · Fish · Trends · Adirondacks

During the early 1990s two studies looked at mercury concentrations in yellow perch from the Adirondack region of New York (Simonin et al. 1994; Driscoll et al. 1994).

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These studies focused on yellow perch because this species is widely distributed, has been studied in other regions and has been found to accumulate relatively high levels of mercury. For example, Simonin et al. (1994) reported mercury concentrations as high as 3.15 µg/g in an Adirondack yellow perch. Both studies found that mercury varied with size and age of the fish and that there was considerable variability in mercury concentrations among fish and among lakes in the Adirondacks. Lakes that were more acidic had higher mercury levels in the fish (Simonin et al. 1994; Driscoll et al. 1994).

Over the past 20 years a number of actions have been undertaken to reduce the availability of mercury in the environment, including air emission controls, recycling mercury, reducing mercury in products and using improved mercury control technologies. These changes will hopefully lead to lower mercury concentrations in fish across New York State. Hammerschmidt and Fitzgerald (2006) recently linked mercury deposition with mercury concentrations in fish. Monitoring of mercury in atmospheric deposition in the Adirondack region of New York was only begun in 2000, but lake sediment cores from 1998 provide a historic record of changes in mercury deposition over time (Raynal et al. 2004). The maximum mercury flux in these cores was observed from 1973 to 1985, and since that time an average decrease of 33% was reported.

The Clean Air Act Amendments of 1990 required reductions in mercury emissions from municipal waste combustors, medical waste incinerators and several other sources. In addition these same regulations required reductions in acidic deposition, and over the past 20 years sulfate deposition in particular has been reduced (Lynch et al. 2000). Recent trends in sulfate concentrations of wet deposition at Huntington Forest in the Adirondack Park show it declining at a rate of 0.84 µeq/l-yr during 1978 to

2004 (Driscoll et al. 2007). Since acidity and sulfate both influence mercury methylation and accumulation by fish (Hrabik and Watras 2002), our expectation is that mercury concentrations in fish should be declining. Natural resource managers, scientists and policy makers are interested in whether this is in fact occurring.

Materials and Methods

This research was started in May 2003 as part of a larger project strategically monitoring mercury in fish from 131 lakes and reservoirs across New York State (Simonin et al. 2008a). Lakes were identified where fish had been previously sampled at least 10 years prior to this project. Fish were collected from 19 lakes during 2003–2005 where fish had been previously analyzed for mercury. Yellow perch were collected from 12 Adirondack lakes that had previously been sampled (Simonin et al. 1994; Driscoll et al. 1994). Where possible we collected 20 yellow perch for the trend analyses and attempted to replicate the same size range of fish as in the earlier studies. Methods of fish collection, handling and measurement of overall length (including caudal fin) were similar to the earlier studies, and fish were analyzed as individual filets. Total mercury was analyzed in the homogenized tissue samples using a modified USEPA Method 1631 (USEPA 2001). Quality control data for mercury were within acceptable limits and included mean values for sample duplicates ($RPD = 3.7 \pm 3.1$), matrix spikes (percent recovery = 99.1 ± 4.8), method blanks (<0.5 ng/g w/w), and certified reference materials (percent recovery = 96.2 ± 3.2). Although the older mercury analyses used slightly different methods, there was no reason to question the validity of the data. Water chemistry in the study lakes was based on samples collected in July and analyzed using methods described in Simonin et al. (2008b).

The New York State Department of Environmental Conservation (NYSDEC) has maintained a database of fish mercury concentrations dating back to 1970, and we selected seven other New York waters not part of the earlier yellow perch studies, where adequate data existed to compare with data from Simonin et al. (2008a). Two of these waters are in the Adirondack Park (Cranberry Lake and Great Sacandaga Lake). Depending on the historical data and the fish collected in the recent survey, we compared mercury concentrations in four fish species: yellow perch, walleye (*Sander vitreus*), largemouth bass (*Micropterus salmoides*) and smallmouth bass (*Micropterus dolomieu*).

In order to compare the fish mercury data among lakes we used a standardized length approach. We determined the overall average size for each fish species in the

2003–2005 dataset, and refer to this as the standard size fish. The standard lengths were 229 mm for yellow perch, 356 mm for smallmouth and largemouth bass, and 457 mm for walleye. For each lake we then used regression to determine the mercury concentration at the standard length (Analytical Software 2003). We used log mercury values to linearize the mercury–fish length relationship. Data were analyzed using analysis of covariance for each lake to determine if there was a significant difference between the mercury–fish length relationship for the older survey with the mercury–fish length relationship in the 2003–2005 data. Fish that were outside of the length range in 1 year were generally excluded from the comparison.

Results and Discussion

For the Adirondack trend analyses we collected an average of 15 yellow perch (range = 7–20 fish) from each of the 12 lakes studied. Using analysis of covariance, half of the lakes showed no significant difference in comparing the older data with the recent data (Table 1). In four of the 12 lakes there was a significant decline in mercury concentrations in the recent fish samples. In two lakes there was a significant increase in mercury concentrations.

When we calculated the mercury concentrations in standard size yellow perch (229 mm) from each lake during the older and newer surveys, we found an overall decrease in mercury concentration in nine out of 12 lakes, with an average decrease of 78 ng/g (95% confidence limits of -189 to $+33$ ng/g; Table 1). Collectively for the 12 lakes, there was a 14% decline (95% confidence limits of -34% to $+6\%$) in mercury concentration in standard size yellow perch. The lake with the lowest mercury concentration in standard size yellow perch was Lake Adirondack, which was also the lake with the lowest mercury concentrations in the study by Simonin et al. (1994). The lakes with higher mercury levels also were the lakes with high levels in earlier studies, although there was some variability. The six lakes which had the highest predicted mercury concentrations in the earlier studies showed lower mercury concentrations in the recent survey (Table 1). Big Moose Lake and Ferris Lake had the largest declines in mercury concentrations in standard size yellow perch.

Water chemistry data collected in the 2003–2005 surveys from the Adirondack study lakes showed lower sulfate levels (28% decrease $\pm 3\%$ SE, $p = 0.0001$) and higher pH (0.35 increase ± 0.08 SE) in all lakes and higher ANC levels in 10 of the 12 lakes when compared to the older samples (Simonin et al. 1994; Blette et al. 1995). The mean increase in ANC was 2.4 $\mu\text{eq/l}$ (± 16.9 SE) and was not statistically significant. Lake Adirondack had a drop in

Table 1 Predicted mercury concentrations (ng/g wet weight) in standard size (229 mm, 9 in.) yellow perch (YP) from Adirondack lakes sampled during 1987 or 1992 compared with 2003–2005 data

Lake	Year	n	Older Hg conc in std size YP	2003–05 n	Recent Hg conc in std size YP	Change in Hg (ng/g)	Significant difference
Adirondack L	87	30	82 (65–104)	20	158 (140–180)	76 (54 to 98)	Increase
Big Moose L	92	25	1,022 (761–1,371)	10	671 (544–828)	–351 (–215 to –486)	Decrease
Chase L	87	26	310 (186–516)	16	588 (505–684)	278 (96 to 459)	ns
Fall L	87	17	488 (376–633)	20	407 (329–505)	–81 (25 to –187)	ns
Ferris L	87	33	841 (496–1,425)	20	580 (490–686)	–261 (–112 to –409)	Decrease
Francis L	92	30	682 (583–799)	10	592 (519–675)	–90 (2 to –183)	ns
Kings Flow	87	28	418 (307–568)	20	232 (185–291)	–186 (–104 to –267)	Decrease
Limekiln L	92	31	579 (142–2,360)	10	347 (224–537)	–232 (418 to –882)	ns
Rondaxe L	92	30	412 (356–478)	10	530 (434–646) ^a	118 (89 to 147)	Increase
Round P	87	28	409 (359–466)	20	392 (359–428)	–17 (23 to –58)	ns
Sunday L	92	30	961 (659–1,401)	18	867 (487–1,542)	–94 (26 to –216)	ns
West Caroga L	87	24	410 (339–496)	7	313 (211–465)	–97 (–56 to –138)	Decrease
Average		27.7	551.2 (374–728)	15.1	473.1 (345–601)	–78.1 (–189 to 33)	Decrease

Data in parenthesis indicate 95% confidence limits. All 1987 data are from Simonin et al. (1994) and 1992 data are from Blette et al. (1995). Significant difference is based on Analysis of Covariance; ns = not significant; $p > 0.05$

^a Fish collected in 2000

ANC of 170 $\mu\text{eq/l}$ during this time period, which had a large influence on the mean change in ANC. The amount of change in water chemistry parameters did not in all cases result in equal changes in mercury in yellow perch, although overall the changes were consistent with expectations. Water chemistry analyses for total and methylmercury were not conducted in all the older survey lakes, so no comparisons were possible.

Seven other New York lakes were studied which had previous fish–mercury data available for comparison with the recent data. The number of fish sampled in the older monitoring efforts was in many cases limited, and required us to combine several years of data in order to produce an adequate number of samples of a particular fish species (Table 2). Largemouth bass trend analysis was possible in Silver Lake and Wappinger Lake, and in both cases showed a significant decrease in mercury concentrations using length as a covariate. Smallmouth bass comparisons were possible in three lakes, Cranberry, Delta and Honeoye. Two of these lakes had significantly lower mercury concentrations in the current study than in the older samples, but Delta Lake had no statistically significant difference in the two groups of data. Yellow perch in Delta Lake showed a small but significant increase in mercury concentrations, while Oneida Lake yellow perch showed no significant change. Walleye trend analysis was possible in Delta, Oneida and Great Sacandaga Lakes. Both Delta Lake and Great Sacandaga Lake walleyes had higher mercury concentrations in the recent survey than in the older data, while Oneida Lake walleyes were not significantly different from

the older data. Water chemistry data were not available for the older fish sample dates, so comparisons of fish mercury levels with water chemistry changes were not possible.

In comparing the 1987–1992 yellow perch mercury data with the 2003–2005 data we observed statistically significant decreases or no change in mercury in fish from most of our study lakes (33% showed a significant decrease, 50% showed no change). We also observed considerable variability in individual lake response. Both of these observations are consistent with other studies (Hrabik and Watras 2002; Hutcheson et al. 2006). In Massachusetts Hutcheson et al. (2006) reported that mercury concentrations in yellow perch from eight lakes decreased an average of 15.4% from 1999 to 2004. Although this was a shorter time period, it compares favorably with our observed average of 14% decrease in mercury concentrations over the past 15–20 years for Adirondack yellow perch. While the Massachusetts study had a greater percentage of lakes with a significant decrease in mercury concentration (62%), the distribution of lakes with a significant decrease in mercury, increase in mercury, or no change in mercury was not significantly different from our lakes ($\chi^2 = 1.71$, $p = 0.42$). Hutcheson et al. (2006) observed larger decreases in mercury in fish (26% to 62% decline) from northeastern Massachusetts waters downwind from an area where mercury emissions from local point sources were substantially reduced. We did not have specific waters where we expected large changes in fish mercury levels due to specific sources of mercury emissions in our study.

Table 2 Predicted mercury concentrations (ng/g wet weight) in standard size yellow perch (YP, 229 mm, 9 in.), smallmouth bass (SMB, 356 mm, 14 in.), largemouth bass (LMB, 356 mm, 14 in.) and walleye (WEYE, 457 mm, 18 in.) from New York State lakes sampled from 1977 to 1994 compared with 2003–2005 data

Lake	Species	Year	n	Hg conc in std size fish	2003–05 n	Recent Hg conc in std size fish	Change in Hg (ng/g)	Significant difference
Cranberry L	SMB	93	11	1,456 (1,271–1,668)	13	842 (689–1,030)	–614 (–458 to –770)	Decrease
Delta L	SMB	88,94	5	477 (388–587)	5	523 (472–578)	46 (–23 to 115)	ns
Delta L	WEYE	88,94	13	605 (513–713)	4	1,059 (498–2,251)	454 (227 to 681)	Increase
Delta L	YP	88,94	8	222 (184–268)	10	276 (218–350)	54 (37 to 71)	Increase
Great Sacandaga L	WEYE	78,82	4	1,236 (959–1,593)	8	1,897 (1,475–2,439)	661 (408 to 914)	Increase
Honeoye L	SMB	83,84	3	274 (186–405)	5	195 (129–296)	–79 (–32 to –126)	Decrease
Oneida L	WEYE	79,81	5	534 (338–843)	10	411 (336–503)	–123 (–296 to 50)	ns
Oneida L	YP	79,81	5	190 (99–365)	10	144 (74–278)	–46 (–98 to 6)	ns
Silver L	LMB	83,90	4	415 (346–497)	10	330 (293–373)	–85 (–60 to –110)	Decrease
Wappinger L	LMB	77,81	5	505 (357–715)	7	379 (268–538)	–126 (–55 to –197)	Decrease

Data in parenthesis indicate 95% confidence limits. The 1977 to 1994 data are from NYSDEC mercury database

Significant difference is based on Analysis of Covariance; ns = not significant; $p > 0.05$)

In an earlier study evaluating trends in contaminants Armstrong and Sloan (1980) reported that mercury concentrations in walleye from Great Sacandaga Lake had increased from 1970 to 1978, and that mercury in largemouth bass from Wappinger Lake had decreased from 1971 to 1977. In our trend analyses for these two waters we found that these same trends have continued. We observed a 53% increase in mercury levels in Great Sacandaga Lake walleyes (457 mm) when we compared the combined 1978/1982 data with the 2005 data (Table 2). In Wappinger Lake we found a 63% decline in mercury concentration in the standard size (356 mm) largemouth bass. Our more recent data are after 20 years time or more, and we cannot say when the changes in mercury occurred. However, it is a concern that the walleye mercury concentrations have continued to increase. Armstrong and Sloan (1980) attributed increases in mercury concentrations in Adirondack waters to increasing acidity due to acidic deposition. However, many Adirondack waters are now becoming less acidic due to reductions in acidic deposition and leading us to expect reductions in fish mercury concentrations. We do not have long-term water chemistry data for Great Sacandaga Lake, but the data from our study indicate that it is neutral in its acidity status (pH = 7.5; Simonin et al. 2008a).

As discussed above, mercury emissions have been reduced substantially in recent years as a result of the Clean Air Act Amendments of 1990. Engstrom and Swain (1997) reported that for Minnesota lakes mercury deposition peaked in the 1960s and 1970s. They also report that global emissions have not been reduced and that decreased inputs to their study lakes were most likely due to regional reductions in emissions. Monitoring of mercury deposition

in the Adirondack region and the northeast in general has been limited, and a recent study was not able to detect a significant reduction in mercury deposition in northeastern North America over the 1996–2002 time-period (VanArsdale et al. 2005). The first year of complete data from the only Adirondack site (Huntington Forest, Hamilton County) was 2000. The lake sediment core data reported by Raynal et al. (2004) are important in showing an average 33% decrease in mercury flux in recent years. Removing mercury from batteries, paint, fungicides and other products has reduced mercury in the environment, and chlor-alkali plants have been replaced by other industrial processes that do not use mercury. However, mercury is a global pollutant, and mercury emissions in other countries, as well as uncontrolled mercury emissions from coal-fired power plants in the US, continue to be substantial.

The variability in the amount of change in fish mercury concentrations among our lakes is certainly in part due to the variability in lake physical and chemical characteristics. There was wide variability among lakes in many of these parameters (Simonin et al. 2008b). In terms of lake chemistry changes over time, Driscoll et al. (2007) showed an approximately 30% decrease in sulfate concentrations in Adirondack lakes over the 1982–2000 time period. When we compared the 2003–2005 sulfate concentrations in our Adirondack yellow perch lakes with water chemistry data collected in either 1987 or 1992, we observed an average decrease of 28%. This reduction in sulfate concentration would likely lead to less methylmercury being available for uptake by the food chain, because it is the sulfate-reducing bacteria that convert mercury into biologically available methylmercury. In a recent study where sulfate was added to a wetland, the result was increased methylmercury

production (Jeremiason et al. 2006). Comparing the two sampling periods in our study, we also found that pH increased in all 12 lakes. We have observed a strong relationship ($r = -0.59$, $p < 0.05$, $n = 103$ lakes) between higher pH levels and lower fish mercury concentrations (Simonin et al. 2008a).

In summary, the downward trends we observed in mercury concentrations in fish were small, but statistically significant and encouraging. Our study demonstrates that mercury is widespread and continues to be at levels of concern in certain lakes, especially in large predatory fish. The connections between mercury emission reductions, changes in mercury deposition, and reductions in fish mercury concentrations are difficult to evaluate and may vary from lake to lake. Larger reductions in emissions and better monitoring of deposition are necessary to document the relationship between emissions and future mercury levels in fish.

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References

- Analytical Software (2003) Statistix 8. Tallahassee, Florida
- Armstrong RW, Sloan RJ (1980) Trends in levels of several known chemical contaminants in fish from New York State waters. New York State Department of Environmental Conservation Technical Report 80-2, Albany, NY
- Blette VL, Munson RK, Gherini SA, Driscoll CT, Schofield CL (1995) Mercury in Adirondack fish and surface waters. Research Report prepared for Empire State Electric Energy Research Corporation, NY
- Driscoll CT, Yan C, Schofield CL, Munson R, Holsapple J (1994) The mercury cycle and fish in the Adirondack lakes. *Environ Sci Technol* 28:136A–143A. doi:10.1021/es00052a003
- Driscoll CT, Driscoll KM, Roy KM, Dukett J (2007) Changes in the chemistry of lakes in the Adirondack region of New York following declines in acidic deposition. *Appl Geochem* 22:1181–1188. doi:10.1016/j.apgeochem.2007.03.009
- Engstrom DR, Swain EB (1997) Recent declines in atmospheric mercury deposition in the Upper Midwest. *Environ Sci Technol* 31:960–967. doi:10.1021/es9600892
- Hammerschmidt CR, Fitzgerald WF (2006) Methylmercury in freshwater fish linked to atmospheric mercury deposition. *Environ Sci Technol* 40:7764–7770. doi:10.1021/es061480i
- Hrabik TR, Watras CJ (2002) Recent declines in mercury concentration in a freshwater fishery: isolating the effects of deacidification and decreased atmospheric mercury deposition in Little Rock Lake. *Sci Total Environ* 297:229–237. doi:10.1016/S0048-9697(02)00138-9
- Hutcheson MS, Rose J, Smith CM, West CR, Pancorbo O, Sullivan J, Eddy B (2006) Massachusetts Fish Tissue Mercury Studies: Long-term Monitoring Results, 1999–2004. Massachusetts Department of Environmental Protection Report, Boston, MA
- Jeremiason JD, Engstrom DR, Swain EB, Nater EA, Johnson BM, Almendinger JE, Monson BA, Kolka RK (2006) Sulfate addition increases methylmercury production in an experimental wetland. *Environ Sci Technol* 40:3800–3806. doi:10.1021/es0524144
- Lynch JA, Bowersox VC, Grimm JW (2000) Changes in sulfate deposition in eastern USA following implementation of Phase I of Title IV of the Clean Air Act Amendments of 1990. *Atmos Environ* 34:1665–1680. doi:10.1016/S1352-2310(99)00426-4
- Raynal DJ, Mitchell MJ, Driscoll CT, Roy KM (2004) Effects of atmospheric deposition of sulfur, nitrogen, and mercury on Adirondack ecosystems. New York State Energy Research and Development Authority, Albany, NY. Report 04-03
- Simonin HA, Gloss SP, Driscoll CT, Schofield CL, Kretser WA, Karcher RW, Symula J (1994) Mercury in yellow perch from Adirondack drainage lakes (New York, U.S.). In: Watras CJ, Huckabee JW (eds) Mercury pollution, integration and synthesis. Lewis Publishers, Boca Raton, Florida, p 457
- Simonin HA, Loukmas JJ, Skinner LC, Roy KM (2008a) Strategic monitoring of mercury in New York State fish. New York State Energy Research and Development Authority, Albany, NY. Report 08-11
- Simonin HA, Loukmas JJ, Skinner LC, Roy KM (2008b) Lake variability: key factors controlling mercury concentrations in New York State fish. *Environ Pollut* 154:107–115. doi:10.1016/j.envpol.2007.12.032
- USEPA (2001) Method 1631, Appendix A, Total mercury in tissue, sludge, sediment, and soil by acid digestion and bromine chloride oxidation. Office of Water, U.S. Environmental Protection Agency (EPA-821-R-01-013)
- VanArsdale A, Weiss J, Keeler G, Miller E, Boulet G, Brulotte R, Poissant L (2005) Patterns of mercury deposition and concentration in Northeastern North America (1996–2002). *Ecotoxicology* 14:37–52. doi:10.1007/s10646-004-6258-x