Ecotoxicology
https://doi.org/10.1007/s10646-019-02119-w

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Spatial patterns and temporal trends in mercury concentrations in common loons (*Gavia immer*) from 1998 to 2016 in New York's Adirondack Park: has this top predator benefitted from mercury emission controls?

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Accepted: 4 October 2019

Abstract

Mercury (Hg), a neurotoxic pollutant, can be transported long distances through the atmosphere and deposited in remote areas, threatening aquatic wildlife through methylation and bioaccumulation. Over the last two decades, air quality management has resulted in decreases in Hg emissions from waste incinerators and coal-fired power plants across North America. The common loon (*Gavia immer*) is an apex predator of the aquatic food web. Long-term monitoring of Hg in loons can help track biological recovery in response to the declines in atmospheric Hg that have been documented in the northeastern USA. To assess spatial patterns and temporal trends in Hg exposure of the common loon in the Adirondack Park of New York State, we analyzed Hg concentrations in loon blood and egg samples from 116 lakes between 1998 and 2016. We found spatially variable Hg concentrations in adult loon blood and feathers across the Park. Loon Hg concentrations (converted to female loon units) increased 5.7% yr^{-1} from 1998 to 2010 ($p = 0.04$), and then stabilized at 1.70 mg kg^{-1} from 2010 to 2016 ($p = 0.91$), based on 760 observations. Concentrations of Hg in juvenile loons also increased in the early part of the record, stabilizing 2 years before Hg concentrations stabilized in adults. For 52 individual lakes with samples from at least 4 different years, loon Hg increased in 34 lakes and decreased in 18 lakes. Overall, we found a delayed recovery of Hg concentrations in loons, despite recent declines in atmospheric Hg.

Keywords Common loon · Adirondack Park · Spatial pattern · Temporal trends · Mercury · New York

Introduction

Mercury (Hg) is a neurotoxic pollutant that accumulates in aquatic food webs, making fish unsafe for consumption by humans and wildlife (Chan et al. 2003; Chen et al. 2008). Mercury emitted from coal-fired power plants, mining, and other sources is transported though the atmosphere (Branch 2008) and deposited in remote areas (Selin et al. 2007), where it can be subsequently converted to methyl-Hg. The transfer of methyl-Hg from lake water and sediments to primary producers is highly efficient, and biomagnification results in increases in tissue Hg concentrations at successive trophic levels (Evers et al. 2007). Wildlife at top trophic levels exposed to Hg can experience adverse behavioral, reproductive, and population-level effects (Driscoll et al. 2013; Eagles-Smith et al. 2018). Remote forested regions have been shown to be particularly sensitive to Hg deposition due to enhanced deposition facilitated by the forest canopy, an abundance of wetlands that are hotspots

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for methylation, and unproductive aquatic ecosystems (Driscoll et al. 2007).

The Clean Air Act of 1990 (https://www.epa.gov/clean-air-act-overview/benefits-and-costs-clean-air-act-1970-1990-retrospective-study) helped regulate Hg emissions to the atmosphere. Additionally, the Mercury and Air Toxics Standards rule (US EPA 2012), which was finalized in 2011, facilitated additional declines in Hg emissions from electric utilities. Anthropogenic Hg emissions declined by 77% from 1990 to 2014 in North America (Zhang et al. 2016) and air Hg concentrations and wet Hg deposition have also decreased across the U.S. and Canada (Weiss-Penzias et al. 2016). In the northeastern U.S., declining regional emissions resulted in declining atmospheric Hg deposition rates from 1992 to 2014 (Gerson and Driscoll 2016; Zhou et al. 2017). Because environmental regulations have been effective in reducing Hg loading, it might be expected that recovery in aquatic ecosystems should follow.

The common loon (*Gavia immer*), a top-trophic predator in the freshwater food web, has long been recognized as an important bioindicator of surface water Hg pollution in remote areas (Evers et al. 1998). The Hg concentrations in loon blood indicate dietary uptake in the current season in the lakes where loons feed (Evers et al. 1998; Burgess and Meyer 2008), as does Hg concentration in loon eggs (Evers et al. 2003). Examining temporal trends in Hg concentrations in loon blood and eggs should therefore provide insight into the efficacy of regional air quality management. In southern New Hampshire, a 64% decrease in mean Hg concentration in loon blood from 1999 to 2002 was attributed to a 45% decrease in upwind Hg emissions from a coal-fired power plant (Evers et al. 2007). To date, long-term trends in loon Hg concentrations have been reported only from Wisconsin (Fevold et al. 2003), partly due to the logistical challenges associated with capturing individual loons repeatedly over multi-year periods.

In addition to temporal patterns, loon Hg concentrations from multiple lakes can indicate spatial variation in Hg exposure. Loon feather Hg concentrations provide insight into Hg exposure over longer time periods, as feathers are the major excretory pathway for Hg sequestration in loons (Evers et al. 1998). Previous studies have reported spatial variation in loon feather and blood Hg concentrations (Evers et al. 1998), primarily related to spatial patterns in lake chemistry (Schoch et al. 2011). For example, acidic lakes methylate Hg more efficiently (Miskimmin et al. 1992; Barkay et al. 1997; Kelly et al. 2003) and thus often contain elevated Hg concentrations in zooplankton, fish (Chen et al. 2005; Adams et al. 2009; Brown et al. 2010), and loons (Meyer et al. 1995; Yu et al. 2011; Schoch et al. 2011). Acid neutralizing capacity (ANC), dissolved organic carbon (DOC), and other physio-chemical variables have been shown to vary with Hg concentrations in aquatic biota across a spatial gradient (Yu et al. 2011). Examining changes in the spatial pattern in loon Hg concentrations should indicate where recovery is accelerating or where it is delayed.

Biodiversity Research Institute, its successor, the Adirondack Center for Loon Conservation, and their collaborators have been monitoring Hg concentrations in common loons in New York's Adirondack Park since 1998, an area previously identified as a biological Hg hotspot (Evers et al. 2007). The dominant source of Hg to the Adirondack Park is atmospheric deposition, estimated to be approximately 20 $\mu g\ m^{-2}\ year^{-1}$ (Miller et al. 2005; Yu et al. 2013). Declines have been reported in lake Hg concentrations and fluxes (Gerson and Driscoll 2016) and in Hg concentrations in yellow perch (*Perca flavescens*) (Simonin et al. 2009; Driscoll et al. this issue). However, spatial patterns in loon Hg concentrations have been explored only for the early part of the record (1998 to 2007; Schoch et al. 2014) and temporal trends have not been examined. Thus, our objectives were to identify spatial patterns and temporal trends in loon Hg concentrations in the Adirondacks using a data set spanning 19 years, 116 lakes, and 760 loon samples. Our goal was to determine whether loon Hg concentrations have decreased in response to reductions in regional and national Hg emissions and deposition.

Methods and materials

Study area and lake selection

Study lakes were located within the Adirondack Park of New York State, USA (43°59′N, 74°14′W), an area of 2.4 million ha. The Adirondack Park is the largest public protected area in the contiguous U.S., containing approximately 2,800 lakes, of which 831 are large enough (>10 ha; Parker 1988) to provide breeding habitat for loons. Study lakes were chosen based on accessibility, observations of loons with juveniles, and the availability of water quality data. Samples of loon blood, eggs and feathers were collected from a subset of study lakes every year, with a total of 116 different lakes sampled from 1998 to 2016. Most lakes (90) were sampled at least twice and the maximum number of years sampled was 11 (4 lakes).

Common loon sampling

We captured common loons for a 1 to 2-week period annually from 1998–2016 using night-lighting and playback techniques (Evers 2001). Upon capture, we collected tissue samples following established protocols (Evers et al. 1998, 2003, 2005). Briefly, we collected loon blood samples from the tibiotarsal vein to determine recent Hg

exposure, and collected feather samples from adult and juvenile loons with fully emerged feathers to provide an indication of long-term Hg accumulation. Feather samples included two central tail feathers and the second secondary feather from each wing. We also banded adult and juvenile (if they were large enough for an adult-sized band) loons with U.S. Geological Survey aluminum bands and a unique combination of plastic colored bands, enabling identification of individual loons to be made from a distance during field observations. In subsequent years, we recorded whether birds had been previously captured, and we collected blood and feather samples from any recaptured loons.

Each breeding season, abandoned loon eggs were opportunistically collected after field staff determined that they were non-viable (i.e., exhibiting a strong odor or abandoned by adult loons). Eggs were processed for Hg following standardized protocols (Evers et al. 2003).

Mercury analysis

Loon blood, feather, and egg samples were submitted to the Animal Health Diagnostics Laboratory, University of Pennsylvania, New Bolton, PA; the Trace Element Research Laboratory, Texas A&M, College Station, Texas; or the Biodiversity Research Institute in Portland, Maine, for analysis of total Hg concentrations. Blood was analyzed as whole blood. Egg samples were freeze dried in a Labconco Lyph Lock 12 freeze dryer and powdered in a Spex Mixer Mill. Blood Hg concentrations are expressed as mg kg^{-1} wet weight, feather Hg concentrations as mg kg^{-1} fresh weight, and egg Hg as mg kg^{-1} wet weight. Egg Hg was not corrected for dehydration; moisture contents averaged 77.1% with a standard deviation of 2.7%. Analyses for methyl-Hg in loon tissues were not conducted because more than 95% of blood Hg in birds occurs in the methyl form (Wolfe et al. 2007).

All tissue samples were analyzed for total Hg concentration either using cold vapor atomic absorption spectroscopy following U.S. Environmental Protection Agency (USEPA) Method 245.1, or using thermal decomposition and atomic absorption spectroscopy (USEPA Method 7473) with an automated direct Hg analyzer. USEPA approved QA/QC protocols were used for all assays (U.S. EPA 1994; U.S. EPA (2007. Recoveries of certified reference materials (DORM-3, DORM-4, DOLT-4, DOLT-5 and BCR464) were >90% and duplicates were within 10%.

Calculation of a common Hg unit

Our dataset included Hg concentrations from male, female, and juvenile loons, as well as loon eggs, all of which naturally differ in Hg concentration. For some analyses, we used the relationships identified by Evers et al. (2011) and adapted by Schoch et al. (2011) to convert all data to female loon units (FLU):

- For adult males, $FLU = e^{(-0.65+1.35\times\ln(\text{male Hg concentration}))}$
- For juveniles < 4 weeks old, $FLU = e^{(1.12+0.44\times\ln(\text{juvenile Hg concentration}))}$
- For juveniles 4–6 weeks old, $FLU = e^{(1.82+0.75\times\ln(\text{juvenile Hg concentration}))}$
- For eggs, $FLU = 0.22 + 1.55 \times$ egg Hg concentration

These conversions allowed us to combine all Hg concentrations from different tissues in a single common metric, which improved the sample size and thus the statistical power.

Analysis of spatial patterns

Spatial patterns of Hg concentration in blood and feathers of male and female loons from 1998 to 2016 were analyzed using Geographic Information System (ESRI ArcGIS 10.3) software. Digital Elevation Model and National Hydrography Dataset of the Adirondack Park were acquired from the New York State GIS Clearinghouse (https://gis.ny.gov/).

To examine whether spatial variation in loon Hg concentrations could be explained by water chemistry, we used previously collected water chemistry data including total Hg, methyl-Hg, pH, ANC, dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), monomeric aluminum (Al_m), and non-labile (organic) monomeric aluminum (Al_o). We had water chemistry data from 2003 and 2004 from 44 of the 116 study lakes (Yu et al. 2011). For these 44 lakes, we averaged FLUs from 1998 to 2016, and used a Pearson correlation test on average FLUs and lake chemistry parameters. We also used a Pearson correlation test on average feather Hg concentration from 1998 to 2016 and 20 lake chemistry parameters, for both male and female loons. This approach ignores change over time in both lakes and loons.

To examine whether feather Hg concentrations could be explained by blood Hg concentrations due to a possible depuration during winter remigial molt, we used a Pearson correlation to test for a correlation between average blood and feather Hg concentrations, using lakes as replicates. The test was conducted for both male and female loons with lakes as observations.

Analysis of temporal trends

We examined temporal trends in loon Hg concentrations (as FLUs) across all of our survey lakes using a general linear mixed model. The mixed model used for trend analysis was $Y_{ij} = x*a + b_i + c_j + u + e_{ij}$, where where i and j are index variables for lake and year, Y_{ij} is the average log-

transformed FLU at lake i in year j, x is a continuous variable for time, a is the slope for temporal trend, b is the coefficient for lake, c is the coefficient for year, u is the intercept, and e_{ij} is the residual error. Restricted maximum likelihood was used to quantify the variance component, and a likelihood ratio test was used to determine the p value. Piecewise regression with the NLIN function was used to identify any changes in slope and to identify a breakpoint between two regressions with different slopes. We log-transformed Hg concentration data to meet the assumption of normality of the residuals.

To examine temporal trends by gender, age, and type of sample (blood vs. egg), we applied the mixed model above to Hg concentrations separately for males, females, juveniles, and eggs.

To analyze temporal trends in loon Hg concentration (as FLUs) in individual lakes, we identified 52 lakes that had sampling intensity ≥ 4 years. We used the nonparametric Mann-Kendall (MK) test along with Sen's method on the annual average FLU. The calculations were performed using the MAKESENS template in Excel (Microsoft Corp., Redmond, WA) which applies the Mann–Kendall test for trends and Sen's slope estimates (Salmi 2002).

We examined the change in blood Hg concentration (not converted to FLUs) of 29 male and 19 female recaptured loons. A general linear model was used to test the fixed effects of gender, number of years between capture and recapture, and lake pH on change in blood Hg concentration. For loons that were captured three or four times (the maximum number of times captured), we used the average Hg concentration of the two closest years as one endpoint, and the number of years between capture and recapture was the difference between the first and last year of capture. We used a t-test to compare the changes in Hg concentration between loons with initial Hg concentrations greater or less than a threshold to test whether Hg concentrations measured at the time of first capture could predict the subsequent changes in Hg concentrations. We selected a moderate Hg exposure risk level (2 mg kg^{-1}, Evers et al. 2008) as the threshold.

All the statistical tests described above were performed using SAS 9.4 (SAS Institute Inc. 2013). We used an α of 0.10 to reduce the chances of accepting the null hypothesis of no effect, and we report the actual p values so that readers can judge the significance of effects (Amrhein et al. 2019).

Results

Loon Hg concentrations from 1998 to 2016

The concentrations of blood Hg in Adirondack common loons from 1998 to 2016 ranged from 0.48–7.31 mg kg^{-1} for males ($\bar{x} = 2.30$ mg kg^{-1}, $n = 153$), 0.11–8.98 mg kg^{-1} for females ($\bar{x} = 2.04$ mg kg^{-1}, $n = 138$), 0.01–0.77 mg kg^{-1} for juveniles < 4 weeks old ($\bar{x} = 0.24$ mg kg^{-1}, $n = 82$), 0.01–0.86 mg kg^{-1} for juveniles from 4 to 6 weeks of age ($\bar{x} = 0.24$ mg kg^{-1}, $n = 64$), and 0.01–1.70 mg kg^{-1} for juveniles > 6 weeks old ($\bar{x} = 0.29$ mg kg^{-1}, $n = 39$). Loon egg Hg concentrations ranged from 0.11–2.65 mg kg^{-1} ($\bar{x} = 0.78$ mg kg^{-1}, $n = 326$; Table 1).

Loon feather Hg concentrations ranged from 3.90–147.23 mg kg^{-1} for males ($\bar{x} = 18.87$ mg kg^{-1}, $n = 151$), and 2.20–35.26 mg kg^{-1} for females ($\bar{x} = 11.23$ mg kg^{-1}, $n = 137$) from 1998 to 2016 (Table 2). Feather Hg concentrations ranged from 0.12–13.44 mg kg^{-1} for juveniles < 6 weeks old ($\bar{x} = 6.40$ mg kg^{-1}, $n = 18$) from 2008 to 2016.

Spatial patterns of loon Hg concentrations

Concentrations of Hg in blood and feathers of male and female loons varied across the Adirondack study area (Fig. 1). Some of the spatial variation in loon blood and egg Hg concentrations was explained by lake chemistry. Higher loon Hg concentrations (using blood and egg data converted to FLUs) were found in lakes with lower pH ($r = -0.44$, $p = 0.002$), lower ANC ($r = -0.34$, $p = 0.05$), higher Al ($r = 0.56$, $p < 0.001$), higher Al_o ($r = 0.48$, $p = 0.004$), higher water methyl-Hg ($r = 0.40$, $p = 0.03$) or higher water total Hg ($r = 0.43$, $p = 0.01$).

Spatial variation of feather Hg for male and female loons was also partially explained by lake chemistry. Lakes with higher Al ($r = 0.31$, $p = 0.08$) had higher Hg concentrations in male loon feathers. Lake with higher Al ($r = 0.41$, $p = 0.03$), Al_o ($r = 0.35$, $p = 0.06$), or methyl-Hg ($r = 0.47$, $p = 0.02$) had higher Hg concentrations in female loon feathers.

We also found that lakes containing loons with higher blood Hg concentrations also had higher feather Hg concentrations for both male ($r = 0.34$, $p = 0.01$) and female loons ($r = 0.52$, $p < 0.01$).

Temporal trends in loon Hg concentrations

Using 760 samples of loon blood and eggs from 116 lakes in the Adirondack Park, we found a significant increase in loon Hg concentrations (as FLU) from 1998 to 2010 ($p = 0.04$), but no significant change from 2010 to 2016 ($p = 0.91$) (Fig. 2). From 1998 to 2010, the annual increase in FLU was 5.7% (0.05 mg kg^{-1} Hg yr^{-1}), while from 2010 to 2016, the FLUs remained stable, with an average of 1.70 mg kg^{-1}. Blood Hg concentration in both male and female loons increased from 1998 to 2010 (males: $p = 0.09$, $n = 153$; females: $p = 0.02$, $n = 138$), but did not change from 2010 to 2016 (both $p \geq 0.25$) (Fig. 3). For juveniles, blood Hg concentration increased from 1998 to 2008 ($p =$

Table 1 Average concentrations of Hg (mg kg^{-1}) in Adirondack loon blood and eggs from 1998 to 2016, with the number of lakes sampled each year (N) and the standard deviation (SD)

Year	Male loons		Female loons		Juveniles < 4 weeks old		Juveniles 4-6 weeks old		Juveniles > 6 weeks old		Loon eggs[a]	
	Ave ± SD	N	Ave ± SD	N	Ave ± SD	N	Ave ± SD	N	Ave ± SD	N	Ave ± SD	N
1998	1.20 ± 0.56	7	0.99 ± 1.11	9	0.01 ± 0.01	7	0.01 ± 0	3	0.04 ± 0.03	2	0.55 ± 0.17	7
1999	3.03 ± 0.94	14	2.18 ± 0.84	10	0.25 ± 0.18	4	0.28 ± 0.27	5	0.49	1	N/A	0
2000	1.72 ± 1.13	8	0.82 ± 0.58	10	0.11 ± 0.13	5	0.07 ± 0.07	9	0.10 ± 0.13	3	0.61 ± 0.37	5
2001	1.94 ± 1.04	10	1.12 ± 0.63	5	0.28 ± 0.08	6	0.25 ± 0	6	0.33 ± 0.14	3	0.58 ± 0.29	17
2002	2.22 ± 0.82	20	1.52 ± 0.66	17	0.18 ± 0.06	8	0.25 ± 0.13	3	0.28 ± 0.09	2	0.72 ± 0.54	15
2003	2.85 ± 0.84	4	2.08 ± 2.28	5	0.38 ± 0.24	5	0.28 ± 0.21	9	0.40 ± 0.18	5	0.67 ± 0.34	9
2004	1.74 ± 1.03	18	1.29 ± 0.86	17	0.23 ± 0.23	11	0.16 ± 0.04	6	0.10	1	0.84 ± 0.63	14
2005	2.77 ± 0.25	2	1.63 ± 0.58	4	0.19 ± 0.05	2	0.21	1	0.22 ± 0.01	2	1.00 ± 0.44	23
2006	1.84 ± 0.57	6	1.35 ± 0.74	6	0.14	1	N/A	0	N/A	0	0.55 ± 0.19	32
2007	2.57 ± 0.96	12	2.21 ± 1.08	10	0.37	1	0.21	1	N/A	0	0.72 ± 0.54	17
2008	2.92 ± 1.71	5	1.81 ± 0.87	8	0.22 ± 0.10	3	0.36 ± 0.27	2	N/A	0	0.55 ± 0.24	15
2009	3.31 ± 2.14	6	3.81 ± 2.98	6	N/A	0	0.66 ± 0.28	3	0.32	1	1.07 ± 0.51	19
2010	1.97 ± 0.84	7	1.75 ± 0.66	5	N/A	0	0.19 ± 0.03	2	N/A	0	1.16 ± 0.58	24
2011	3.07 ± 2.08	7	1.86 ± 0.69	5	0.19 ± 0.12	6	0.28 ± 0.03	2	0.36 ± 0.09	6	1.11 ± 0.68	19
2012	2.63 ± 0.61	4	1.96 ± 1.15	5	0.22 ± 0.04	3	0.27 ± 0.15	3	0.25 ± 0.02	2	0.55 ± 0.18	13
2013	2.09 ± 0.91	5	1.26 ± 0.30	3	0.23 ± 0.04	5	N/A	0	0.24 ± 0.04	3	0.74 ± 0.24	19
2014	2.42 ± 1.12	11	2.04 ± 1.15	7	0.24 ± 0.10	11	0.10 ± 0.04	3	N/A	0	0.86 ± 0.30	20
2015	0.72	1	7.25	1	0.72	1	0.35 ± 0.32	3	0.59 ± 0.64	6	0.68 ± 0.53	29
2016	2.78 ± 0.84	6	1.91 ± 0.42	5	0.14 ± 0.05	3	0.22 ± 0.04	3	0.29 ± 0.14	2	0.89 ± 0.31	29

[a]Concentrations of egg Hg as wet weight in 2009, 2010 and 2011 were calculated based on the Hg concentration as dry weight and an average moisture content of 294 eggs measured in other years.

Concentrations of Hg are reported as wet weight. N/A indicates that data are not available.

0.03), and did not change from 2008 to 2016 ($p = 0.38$). For eggs, Hg concentration increased from 1998 to 2011 ($p = 0.01$) and did not change from 2011 to 2016 ($p = 0.49$) (Fig. 3). These concentrations can also be viewed in risk categories from low to extra high (Fig. 4)

In addition to testing for a temporal trend across the entire data set, we characterized change over time within individual lakes; there were 52 lakes with at least 4 years of loon Hg samples. Patterns within these lakes were not consistent ($p = 0.51$ for a t-test on slopes of FLUs over time). Only eight lakes had statistically significant rates of change, with six lakes showing increases and two lakes showing decreases (Table 3).

Changes of Hg concentrations in blood from recaptured loons

Female loons had larger differences in Hg concentrations between their first and last capture than did male loons ($p = 0.09$ for ANOVA comparing changes in female vs. male loons). Neither the period between capture and recapture ($p = 0.45$) nor lake pH ($p = 0.34$) explained changes in Hg concentrations between sampling dates.

Concentration of loon blood Hg measured at the time of first capture was an important factor influencing the subsequent changes in Hg concentration. Loons with a lower initial blood Hg concentration (<2 mg kg^{-1}) tended to have higher Hg concentrations at their subsequent recapture (25 loons had increases and 4 loons had decreases; Fig. 5a). Loons with a higher initial blood Hg concentration (>2 mg kg^{-1}) had lower concentrations upon recapture (Fig. 5b), with only two exceptions. One of these two loons was captured in two different lakes, with the second lake having higher Hg concentrations in zooplankton and fish than the initial lake. The other loon was from a lake with one of the highest total Hg concentrations (3.8 ng L^{-1}) and the lowest pH (5.2) in the water column of the lakes studied.

Discussion

Loon Hg concentrations

In this study of Adirondack common loons, average male blood Hg concentrations (2.30 ± 0.09 mg Hg kg^{-1}) were in the moderate-high Hg risk exposure category for loons

Table 2 Average concentrations of Hg (mg kg^{-1}) in Adirondack loon feathers from 1998 to 2016, with the number of lakes sampled each year (N) and the standard deviation (SD)

Year	Male		Female	
	Ave ± SD		Ave ± SD	N
1998	18.70 ± 8.22	7	9.46 ± 4.09	9
1999	14.87 ± 7.16	14	10.56 ± 3.34	10
2000	8.97 ± 2.22	8	8.70 ± 2.21	10
2001	18.19 ± 8.43	10	10.22 ± 6.82	5
2002	28.54 ± 29.29	20	15.55 ± 8.77	17
2003	20.63 ± 5.33	4	11.22 ± 4.04	5
2004	18.10 ± 11.43	18	10.08 ± 4.70	17
2005	20.70 ± 4.96	2	15.33 ± 4.26	4
2006	32.98 ± 8.09	5	12.42 ± 4.12	5
2007	17.68 ± 5.55	12	11.02 ± 3.32	10
2008	11.26 ± 9.65	5	7.47 ± 2.41	8
2009	19.01 ± 9.97	7	12.22 ± 3.43	5
2010	12.43 ± 5.73	7	11.70 ± 4.32	5
2011	13.71 ± 5.97	7	8.68 ± 3.96	5
2012	17.85 ± 6.58	4	10.86 ± 2.51	5
2013	16.44 ± 4.14	5	10.80 ± 5.07	3
2014	20.17 ± 12.58	10	12.96 ± 7.96	7
2015	12.60	1	26.25	1
2016	22.09 ± 20.30	6	9.02 ± 1.42	5

Concentrations of Hg are reported as fresh weight. Feather Hg concentrations in juveniles are not reported because birds were sampled for only 4 years in 16 lakes

(2–3 mg Hg kg^{-1}; Evers et al. 2008) and average female loon blood Hg levels (2.04 ± 0.11 mg Hg kg^{-1}) were at the low end of the moderate-high Hg risk exposure category. Lower blood Hg concentrations of female loons compared to males can be explained by the excretion of Hg into eggs during the breeding season and the ingestion of smaller prey that contain lower quantities of Hg. The Hg concentration in loon eggs averaged 0.78 ± 0.05 mg kg^{-1} from 1998 to 2016, which is categorized as moderate risk for loon exposure (0.5–1.3 mg Hg kg^{-1}; Evers et al. 2003).

Blood Hg is indicative of recent dietary exposure, whereas feather Hg is reflective of longer-term exposure (Evers et al. 1998; Burgess and Meyer 2008). Feather Hg concentrations averaged 18.2 ± 1.2 mg kg^{-1} for males and 10.8 ± 0.5 mg kg^{-1} for females, with only 3% of male and 0% of female loons exceeding the adverse effects threshold of 40 mg Hg kg^{-1} (Evers et al. 2008). Thus, fewer of the loons studied exceeded the adverse effects category based on feather Hg concentrations, compared to the moderate risk of Hg exposure based on blood and egg Hg concentrations. This might suggest that the summer diet of loons in the Adirondacks contains more Hg than does their diet in their wintering areas. However, feather and blood Hg concentrations were correlated, as has been reported elsewhere in North America using data from 1992 to 1996 (Evers et al. 1998), which suggests that differences in winter diet are not very important to Hg accumulation in feathers.

We note that loon capture success is skewed toward those birds who are raising young. Loons with the highest Hg levels are less likely to hatch chicks successfully (Evers et al. 2008), and thus are less likely to be included in this dataset. This omission suggests that the Hg levels we report, based on captured loons, likely underestimate Hg exposure and its impacts on the health of the overall loon population. However, since this bias is consistent over time, it does not preclude the detection of long-term trends in Hg in the loon population in the Adirondack Park.

Spatial pattern in loon Hg

The relationship between loon Hg concentration and lake chemistry (e.g., pH, ANC) was similar to that reported for the early years of observation (Schoch et al. 2014). In lakes with lower pH, methylation rates are hypothesized to be higher because there is less dissolved organic carbon to complex Hg^{2+}, making Hg^{2+} more available for methylation (Miskimmin et al. 1992; Barkay et al. 1997; Kelly et al. 2003). Thus, more acidic lakes could have higher Hg methylation resulting in higher bioaccumulation and biomagnification rates (Steffan et al. 1988; Kelly et al. 2003; Yu et al. 2011). Note that Yu et al. (2011) found no relation between methyl-Hg concentrations with pH in the water column of Adirondack lakes, but observed increases in the methyl-Hg concentrations in zooplankton of those same lakes, and this pattern of elevated Hg with lower pH persisted up the aquatic food chain. This pattern led these investigators to suggest that bioaccumulation of methyl-Hg at the base of the food chain increased with decreases in pH, and this factor controlled the spatial relationship of fish and loon Hg with pH and ANC. Other landscape characteristics including elevation, watershed area (Yu et al. 2011) and land cover (Kramar et al. 2005) might also affect the spatial distribution of loon Hg concentrations.

Temporal trends in loon Hg

We expected to observe declines in Hg concentrations in Adirondack loons over the course of this long-term study, reflecting the reported declines in atmospheric dry (Zhou et al. 2017) and wet (Mao et al. 2017) Hg deposition, lake water Hg concentrations and fluxes (Gerson and Driscoll 2016; Millard et al. this issue), and Hg concentrations in yellow perch (Simonin et al. 2009; Driscoll et al. this issue). However, over the full dataset of 116 lakes, we found that loon Hg concentrations (based on FLUs) increased from 1998 to 2010 and then remained stable from 2011–2016.

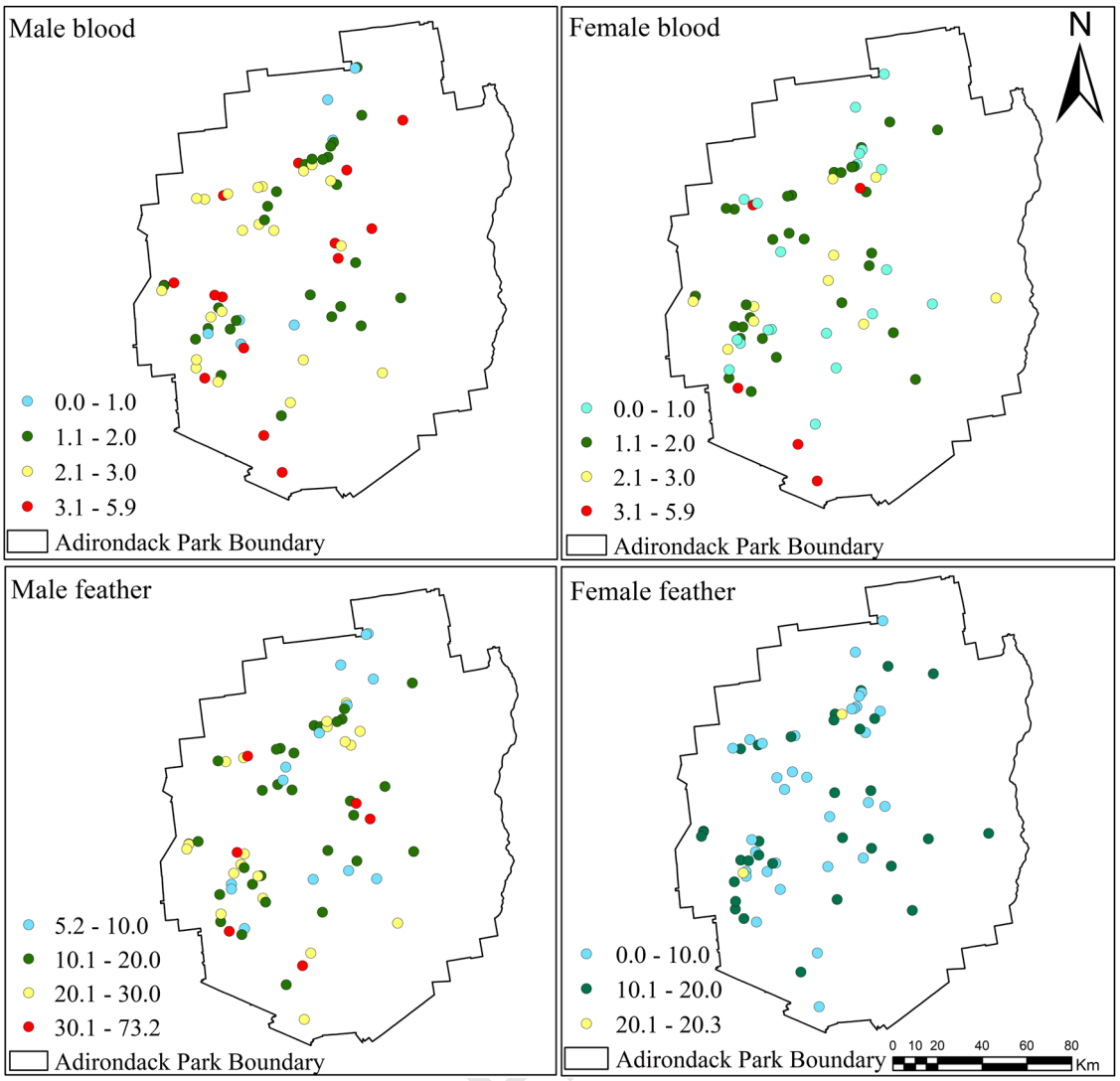

Fig. 1 Spatial patterns of Hg concentrations in blood and feathers of male and female loons in the Adirondack Park from 1998–2016

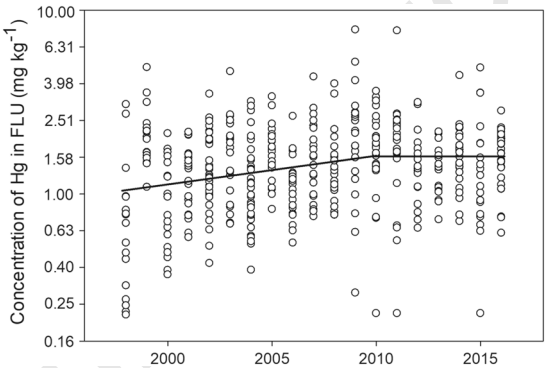

Fig. 2 Temporal trends in Hg concentrations, as FLUs, in Adirondack loons from 1998 to 2016

Juvenile blood Hg concentration stabilized earlier than adults. Adult loons consume older and larger fish that have accumulated more Hg (Driscoll et al. 1994), while juvenile loons consume younger and smaller fish that benefit sooner from lower Hg concentrations in the environment (Driscoll et al. this issue). The delay in recovery of loon Hg compared to fish Hg is likely be associated with the higher trophic position of loons in the aquatic ecosystem, as in the case of bald eagles in New York State, which had high Hg in 2004–2006 (DeSorbo et al. 2019, this issue).

Even recaptured adult loons in the Adirondacks did not show a consistent temporal change in Hg concentration (Fig. 5). The fact that recaptured loons showed changes towards a central tendency might be due to random variation, as a random high value would be unlikely to be followed by a higher value. Near Little Rock Lake, Wisconsin, where atmospheric Hg deposition declined 10% annually from 1994 to 2000 (Hrabik and Watras 2002), recaptured adult loons showed detectable improvement (5% yr^{-1}) in blood Hg from 1992 to 2000 (Fevold et al. 2003). The

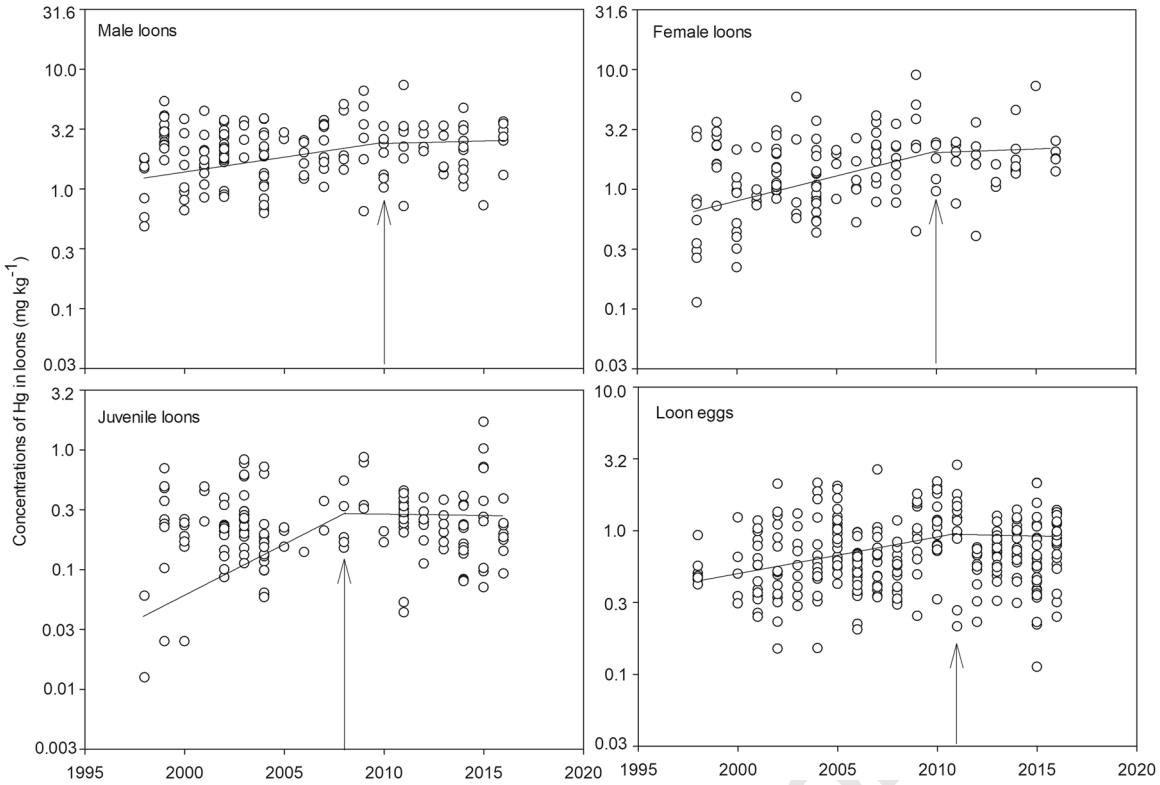

Fig. 3 Temporal trends in Hg concentrations, in the native units, in male, female, and juvenile loons and loon eggs from lakes in the Adirondacks from 1998 to 2016. Arrows indicate the breakpoints in piecewise regression

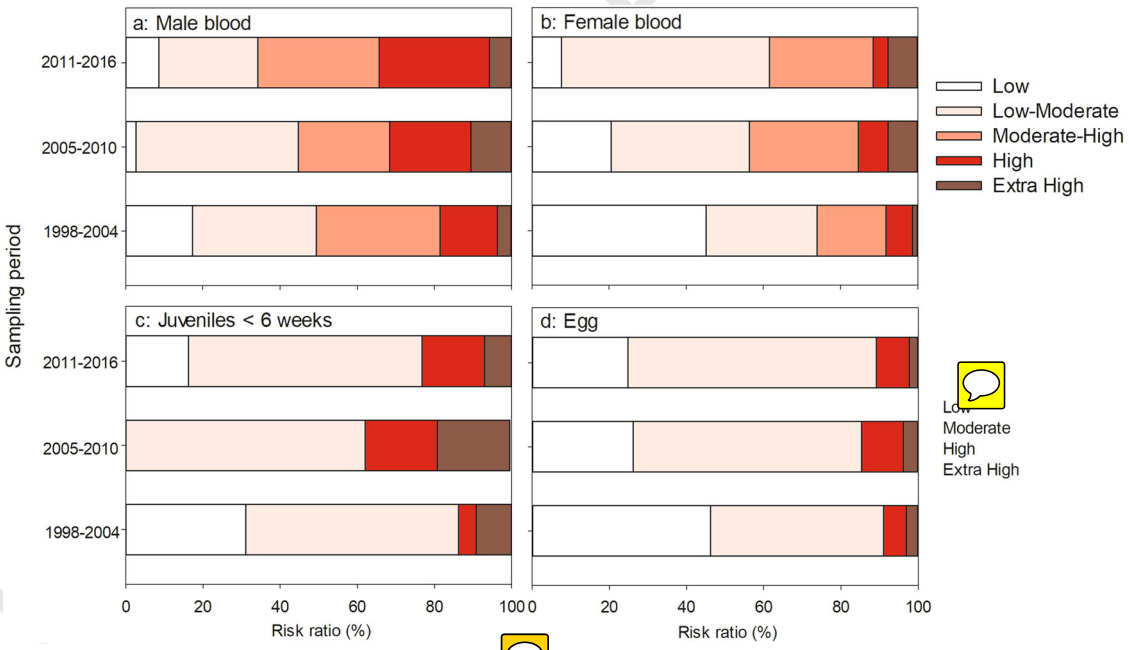

Fig. 4 Risk ratios for Hg exposure based on Hg exposure groups: (0–1 mg kg^{-1}), low-moderate (1–2 mg kg^{-1}), moderate-high (2–3 mg kg^{-1}), high (3–4 mg kg^{-1}) and extra high (> 4 mg kg^{-1}) for ~~Male~~ **a** and female blood **b** (Evers et al. 2008, Burgess and Meyer 2008), low (0–0.1 mg kg^{-1}), moderate (0.1–0.3 mg kg^{-1}), high (0.3–0.4 mg kg^{-1}) and extra high (>0.4 mg kg^{-1}) for juveniles < 6 weeks **c** (Meyer et al. 1998), and low (0–0.5 mg kg^{-1}), moderate (0.5–1.3 mg kg^{-1}), high (1.3–2.0 mg kg^{-1}) and extra high (>2.0 mg kg^{-1}) for eggs **d** (Evers et al. 2003)

Table 3 Temporal trends in loon Hg concentrations (as FLUs) at each lake that was sampled ≥ 4 years, using nonparametric Mann-Kendall with Sen's slope estimates

Watershed	Lake	Initial year	Final year	Initial FLU	Final FLU	ears sampled	Trend slope
Black	Beaver Lake-Lewis Cty	1999	2007	3.55	2.35	6	−0.21
	Big Moose Lake	2000	2016	2.13	2.26	10	−0.04
	Dart Lake	2002	2010	2	0.22	5	−0.24[a]
	First Lake-Fulton Chain	2001	2016	1.13	1.12	4	0.01
	Limekiln Lake	1999	2016	1.48	2.17	11	0.01
	Little Moose Lake	2009	2015	1.32	0.75	5	−0.12
	Little Safford Lake	1998	2010	0.23	3.02	5	0.24[a]
	Moshier Reservoir	1999	2005	2.21	3.03	4	0.17
	Moss Lake	1999	2016	1.57	2	11	0.02
	Nicks Lake	1998	2016	3.07	1.94	6	−0.05
	North Lake	1998	2015	2.72	4.86	5	0.15
	Rondaxe Lake	1998	2011	0.82	2.27	4	0.12[b]
	Sixth Lake	2002	2016	0.52	0.61	4	0.02
	South Lake	1998	2013	0.77	1.45	8	0.09
	Squaw Lake‘	2001	2016	0.76	1.22	4	0.01
	Twitchell Lake	1999	2016	3.15	1.87	7	−0.08
	Woodhull Lake	2000	2016	1.48	2.11	6	−0.03
Champlain	Lake Kushaqua	2004	2016	1.08	1.98	7	0.07[b]
	Little Clear Pond	1998	2015	0.27	0.68	11	0.01
	Long Pond-Franklin Cty	2000	2016	1.24	2.29	9	0.03
	Middle Pond	2002	2016	2.49	2.05	5	−0.02
	Middle Saranac Lake	2002	2014	1.1	1.39	8	0.05
	Putnam Pond	2000	2014	0.71	1.84	5	0.09
	Silver Lake-Clinton Cty	2008	2016	3.98	1.66	5	−0.18
	Taylor Pond	2002	2016	2.16	1.66	7	−0.02
	Weller Pond	1998	2012	0.25	3.14	4	0.17
Mohawk	Canada Lake	2003	2011	1.98	2.13	5	0.08
	Ferris Lake	1999	2006	4.89	1.73	6	−0.44[a]
	G Lake	2003	2012	2.03	1.65	4	−0.08
Oswegatchie	Chaumont Pond	2001	2016	1.37	1.75	7	0.03
	Cranberry Lake	1999	2015	2.42	3.54	10	0.12
Raquette	Hitchins Pond	1999	2016	2.17	1.06	7	−0.02
	Lows Lake	1999	2015	1.67	0.63	8	−0.05
	Piercefield Flow	2004	2014	0.78	1.02	4	0.09
St. Lawrence	Massawepie Lake	2001	2016	0.88	1.52	6	0.10
St. Lawrence-Canada	Clear Pond	2002	2016	0.42	0.74	5	−0.002
	Dry Channel Pond	2002	2016	2.6	2.83	4	0.03
	Lower Saint Regis Lake	2012	2016	1.76	1.39	4	−0.16
	Spitfire Lake	1998	2010	0.48	2.05	6	0.13[a]
	Upper Saint Regis Lake	1998	2014	0.22	1.31	8	0.02
Upper Hudson	Arbutus Pond	1998	2014	0.97	1.8	10	0.03
	Catlin Lake	2008	2013	1.27	1.57	4	0.21
	Deer Pond	1998	2014	1.38	0.71	6	−0.04
	Garnet Lake	2002	2016	1.26	1.91	5	0.04
	Henderson Lake	2003	2016	2.07	2.01	7	−0.004
	Lake Abanakee	2002	2015	0.64	1.01	5	0.03[b]
	Lake Durant	1999	2009	1.99	2.2	5	0.008
	Mason Lake	1999	2014	1.62	1.68	5	−0.03
	Oliver Pond	2002	2016	0.98	1.07	4	0.001
	Piseco Lake	2001	2012	1.63	1.3	6	0.02
	Wolf Pond	1998	2016	0.69	1.87	11	0.05[a]
	Woodruff Pond	2004	2012	0.55	1.41	3	0.11

[a]indicated slopes significant at $\alpha = 0.05$, and [b]indicated slopes significant at $\alpha = 0.10$

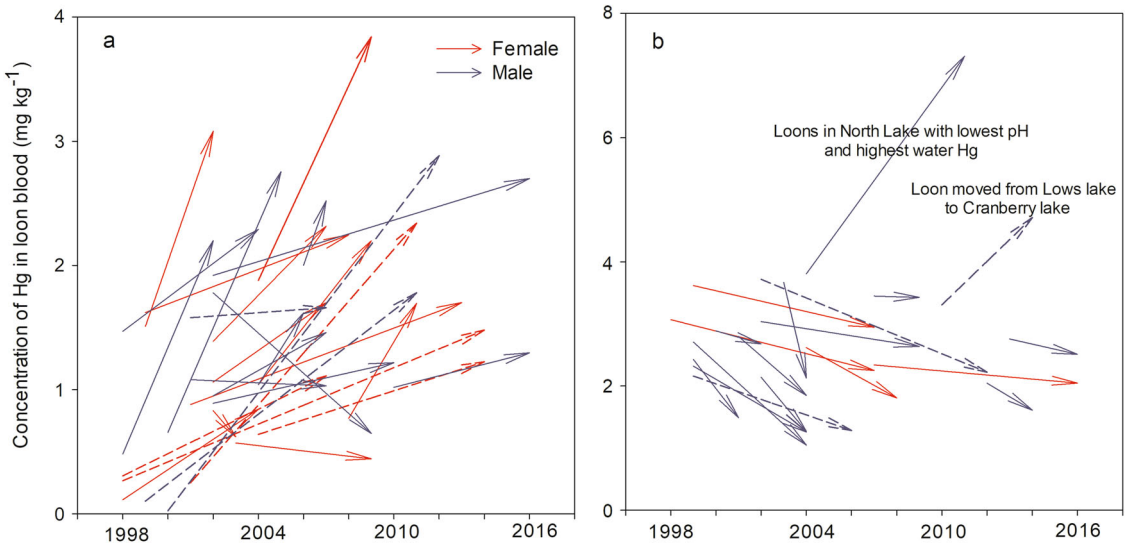

Fig. 5 Temporal changes in Hg concentrations in loon blood from recaptured Adirondack loons with an initial Hg concentration **a** <2 mg kg^{-1} and **b** >2 mg kg^{-1}. Of the 48 recaptured loons, 4 female loons and 6 male loons were captured and recaptured at different lakes (dashed arrows)

modest reductions of atmospheric Hg deposition in our study area (1.6% annually; Zhou et al. 2017) have yet to produce consistently decreasing loon Hg concentrations either for the entire data set or for recaptured loons.

Implications for loon recovery

It is difficult to predict the recovery of loon Hg concentrations. Our original hypothesis was that loon Hg in the Adirondack Park of New York would decrease in response to decreases in regional Hg emissions and deposition. Although the rate of increase in loon Hg has attenuated in recent years, there is no evidence of recovery from elevated exposure. In addition to decreases in Hg deposition, the Adirondack Park is experiencing decreases in acid deposition (Driscoll et al. 2016) and changing climate (USGCRP, 2018), both of which can alter Hg dynamics in complex ways. Decreases in acid deposition have resulted in decreases in sulfate and increases in pH and ANC, which should decrease Hg methylation and bioaccumulation. However, decreases in acid deposition have also coincided with increases in dissolved organic matter (Driscoll et al. 2016), which could drive increases in Hg methylation and have been shown to correspond with increases in fish Hg in Scandinavia (Hongve et al. 2012). In the Adirondacks and the northeastern U.S., air temperature and precipitation have also increased (USGCRP, 2018). These changes have altered watershed hydrology through decreases in snowpack accumulation and duration, earlier snowmelt, and changing stratification of lakes (Campbell et al. 2011). Moreover, climate is becoming more variable with more extended dry and wet periods and extreme runoff events. Increases in temperature will tend to increase mineralization and methylation rates. Increases in precipitation and runoff will tend to increase Hg transport from watersheds to aquatic ecosystems. Increases in wet-dry cycles and stratification will enhance methylation (Evers et al. 2007). In total, these climatic effects will likely increase forest watershed sensitivity to atmospheric Hg deposition. Although these hydrologic and biogeochemical effects have been, and undoubtedly will continue to be, profound, the greatest uncertainty in changes in Hg dynamics may be those manifested though changes in biological communities. Changes in acid deposition, climate, species invasion, and other drivers are changing aquatic biological communities in complex ways. Changes in species distribution, biomass, and food chains can have profound and unpredictable effects on Hg concentrations in apex predators.

In conclusion, although Hg concentrations in loon blood, eggs, and feathers have yet to show recovery, the lack of increase in the last decade suggests that loons have benefited from controls on Hg emissions. It will be difficult to predict the ultimate recovery of loon populations because Hg dynamics are influenced by a host of ecosystem biophysical processes, as described above. Given this complexity, it will be important to continue long-term measurements of loon Hg with careful attention to monitoring design (Yang et al. this issue). Interpretation could be facilitated if loon Hg measurements were accompanied by fish and water chemistry observations.

Acknowledgements We greatly appreciate the many hours the Adirondack field staff have devoted each summer to capturing, color-banding, and sampling loons. The Adirondack Watershed Institute of Paul Smiths College and the Adirondack Ecological Center of SUNY ESF generously provided students annually to assist with monitoring loons on some of our study lakes. The New York State Department of Environmental Conservation, the Wildlife Conservation Society's Zoological Health Program, and Calvin College have provided in-kind support, staff, and equipment for the loon capture and sampling fieldwork each year.

Funding Financial support was provided by the New York State Energy Research and Development Authority, the Wildlife Conservation Society, The Wild Center, the Raquette River Advisory Council, and numerous private foundations and donors.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval All applicable international, national, and institutional guidelines for the care and use of animals were followed. All procedures performed in studies involving animals were in accordance with the ethical standards of the institution or practice at which the studies were conducted.

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References

Adams RM, Twiss MR, Driscoll CT (2009) Patterns of mercury accumulation among seston in lakes of the Adirondack Mountains, New York. Environ Sci Technol 43:4836–4842

Amrhein V, Greenland S, McShane B (2019) Scientists rise up against statistical significance. Nature 567:305–307

Barkay T, Gillman M, Turner RR (1997) Effects of dissolved organic carbon and salinity on bioavailability of mercury. Appl Environ Microbiol 63:4267–4271

Branch UC (2008) The global atmospheric mercury assessment: sources, emissions and transport. UNEP-Chemicals, Geneva

Brown D, Goncharov A, Paul E, Simonin H, Carpenter DO (2010) The relationship between Adirondack lake pH and levels of mercury in yellow perch. J Aquat Anim Health 22:280–290

Burgess NM, Meyer MW (2008) Methylmercury exposure associated with reduced productivity in common loons. Ecotoxicology 17:83–91

Campbell JL, Driscoll CT, Pourmokhtarian A, Hayhoe K (2011) Streamflow responses to past and projected future changes in climate at the Hubbard Brook Experimental Forest, New Hampshire, United States. Water Resour Res 47:W02514. https://doi.org/10.1029/2010WR009438

Chan HM, Scheuhammer AM, Ferran A, Loupelle C, Holloway J, Weech S (2003) Impacts of mercury on freshwater fish-eating wildlife and humans. Hum Ecol Risk Assess 9:867–883

Chen CY, Stemberger RS, Kamman NC, Mayes BM, Folt CL (2005) Patterns of Hg bioaccumulation and transfer in aquatic food webs across multi-lake studies in the northeast US. Ecotoxicology 14:135–147

Chen C, Amirbahman A, Fisher N, Harding G, Lamborg C, Nacci D, Taylor D (2008) Methylmercury in marine ecosystems: spatial patterns and processes of production, bioaccumulation, and biomagnification. EcoHealth 5:399–408

Desorbo CR, Burgess NM, Nye P, Loukmas JJ, Brant HA, Burton M, Persico CP, Evers DC (2019) Mercury exposure differences in nestling bald eagles in New York, USA. Ecotoxicology. (in review, this issue)

Driscoll CT, Yan C, Schofield CL, Munson R, Holsapple J (1994) The mercury cycle and fish in the Adirondack lakes. Environ Sci Technol 28(3):136A–143A

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

Driscoll CT, Mason RP, Chan HM, Jacob DJ, Pirrone N (2013) Mercury as a global pollutant: sources, pathways, and effects. Environ Sci Technol 47(10):4967–4983

Driscoll CT, Driscoll KM, Fakhraei H, Civerolo K (2016) Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmos Environ 146:5–14. https://doi.org/10.1016/j.atmosenv.2016.08.034

Driscoll CT, Gerson JR, Taylor MS, Millard GM, Shaw AL, Yang Y, Paul EA (2019) Patterns and trends in water and fish mercury in the Adirondacks. Ecotoxicology. (this issue)

Eagles-Smith CA, Silbergeld EK, Basu N, Bustamante P, Diaz-Barriga F, Hopkins WA, Kidd KA, Nyland JF (2018) Modulators of mercury risk to wildlife and humans in the context of rapid global change. Ambio 47(2):170–197

Evers DC, Kaplan JD, Meyer MW, Reaman PS, Braselton WE, Major A, Burgess N, Scheuhammer AM (1998) Geographic trend in mercury measured in common loon feathers and blood. Environ Toxicol Chem 17:173–183

Evers DC (2001) Common loon population studies: Continental mercury patterns and breeding territory philopatry. Ph.D. dissertation, University of Minnesota, St. Paul, MN

Evers DC, Taylor KM, Major A, Taylor RJ, Poppenga RH, Scheuhammer AM (2003) Common loon eggs as indicators of methylmercury availability in North America. Ecotoxicology 12:69–81

Evers DC, Burgess NM, Champoux L, Hoskins B, Major A, Goodale WM, Taylor RJ, Poppenga R, Daigle T (2005) Patterns and interpretation of mercury exposure in freshwater avian communities in northeastern North America. Ecotoxicology 14:193–221

Evers DC, Han YJ, Driscoll CT, Kamman NC, Goodale MW, Lambert KF, Holsen TM, Chen CY, Clair TA, Butler T (2007) Biological mercury hotspots in the northeastern United States and southeastern Canada. AIBS Bull 57:29–43

Evers DC, Savoy LJ, DeSorbo CR, Yates DE, Hanson W, Taylor KM, Siegel LS, Cooley JH, Bank MS, Major A, Munney K (2008) Adverse effects from environmental mercury loads on breeding common loons. Ecotoxicology 17:69–81

Evers DC, Williams KA, Meyer MW, Scheuhammer AM, Schoch N, Gilbert AT, Siegel L, Taylor RJ, Poppenga R, Perkins CR (2011) Spatial gradients of methylmercury for breeding common loons in the Laurentian Great Lakes region. Ecotoxicology 20:1609–1625

Fevold BM, Meyer MW, Rasmussen PW, Temple SA (2003) Bioaccumulation patterns and temporal trends of mercury exposure in Wisconsin Common Loons. Ecotoxicology 12:83–93

Gerson JR, Driscoll CT (2016) Is mercury in a remote forested watershed of the Adirondack mountains responding to recent decreases in emissions? Environ Sci Technol 50:10943–10950

Hongve D, Haaland S, Riise G, Blakar I, Norton S (2012) Decline of acid rain enhances mercury concentrations in fish. Environ Sci Technol 46(5):2490–2491. https://doi.org/10.1021/es3002629

Hrabik TR, Watras CJ (2002) Recent declines in mercury concentration in a freshwater fishery: isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. Sci Total Environ 297:229–237

Kelly CA, Rudd JW, Holoka MH (2003) Effect of pH on mercury uptake by an aquatic bacterium: implications for Hg cycling. Environ Sci Technol 37:2941–2946

Kramar D, Goodale WM, Kennedy LM, Carstensen LW, Kaur T (2005) Relating land cover characteristics and common loon mercury levels using geographic information systems. Ecotoxicology 14:253–262

Mao H, Ye Z, Driscoll CT (2017) Meteorological effects on Hg wet deposition in a forested site in the Adirondack region of New York during 2000–2015. Atmos Environ 168:90–100

Meyer MW, Evers DC, Daulton T, Braselton WE (1995) Common loons (*Gavia immer*) nesting on low pH lakes in northern Wisconsin have elevated blood mercury content. In: Mercury as a global pollutant. Springer, Netherlands. pp. 871–880

Meyer MW, Evers DC, Hartigan JJ, Rasmussen PS (1998) Patterns of common loon (*Gavia immer*) mercury exposure, reproduction, and survival in Wisconsin, USA. Environ Toxicol Chem: Int J 17:184–190

Millard GD, Driscoll CT, Montesdeoca M, Yang Y, Taylor MS, Boucher S, Shaw AL, Paul EA, Parker C, Yokota K (2019) Patterns and trends of fish mercury in New York State. Ecotoxicology. (this issue)

Miller EK, Vanarsdale A, Keeler GJ, Chalmers A, Poissant L, Kamman NC, Brulotte R (2005) Estimation and mapping of wet and dry mercury deposition across northeastern North America. Ecotoxicology 14:53–70

Miskimmin BM, Rudd JWM, Kelly CA (1992) Influence of dissolved organic carbon, and microbial respiration rates on mercury methylation and demethylation in lake water. Can J Fish Aquat Sci 49:17–22

Parker KE (1988) Common loon reproduction and chick feeding on acidified lakes in the Adirondack Park, New York. Can J Zool 66:804–810

Salmi T (2002) Detecting trends of annual values of atmospheric pollutants by the Mann–Kendall test and Sen's slope estimates-the Excel template application MAKESENS. ~~Ilmatieteen laitos,~~

SAS Institute Inc. (2013) SAS 9.4 guide to software. Updates. SAS Institute Inc, Raleigh, North Carolina

Schoch N, Glennon M, Evers D, Duron M, Jackson A, Driscoll C, Yu X, Simonin H (2011) Long-term monitoring and assessment of mercury based on integrated sampling efforts using the common loon, prey fish, water, and sediment. NYSERDA Report No. 12–06: 116. https://www.nyserda.ny.gov/About/Publications/Research-and-Development-Technical-Reports/Environmental-Research-and-Development-Technical-Reports#eco. Accessed 25 Jan 2017

Schoch N, Glennon MJ, Evers DC, Duron M, Jackson AK, Driscoll CT, Ozard JW, Sauer AK (2014) The impact of mercury exposure on the Common Loon (*Gavia immer*) population in the Adirondack Park, New York, USA. Waterbirds 37:133–146

Selin NE, et al. (2007) Chemical cycling and deposition of atmospheric mercury: global constraints from observations. J Geophys Res Atmos 112(D2)

Simonin HA, Loukmas JJ, Skinner LC, Roy KM, Paul EA (2009) Trends in mercury concentrations in New York State fish. Bull Environ Contamination Toxicol 83(2):214–218

Steffan RJ, Korthals ET, Winfrey MR (1988) Effects of acidification on mercury methylation, demethylation, and volatilization in sediments from an acid-susceptible lake. Appl Environ Microbiol 54:2003–2009

U.S. EPA (1994) Method 245.1: determination of mercury in water by cold vapor atomic absorption spectrometry. Revision 3.0. U.S. EPA, Cincinnati, OH

U.S. EPA (2007) Mercury in solids and solutions by thermal decomposition, amalgamation, and atomic absorption spectrophotometry. EPA-7473. U.S. EPA. p 17

U.S. EPA (2012). Mercury and air toxics standards for power plants. 40 CFR parts 60 and 63. U.S. EPA. https://www.gpo.gov/fdsys/pkg/FR-2012-02-16/pdf/2012-806.pdf

USGCRP (2018) In: Reidmiller, DR, CW Avery, DR Easterling, KE Kunkel, KLM Lewis, TK Maycock, and BC Stewart (eds) Impacts, risks, and adaptation in the United States: Fourth National Climate Assessment, Volume II: Report-in-Brief. U.S. Global Change Research Program, Washington, DC, USA, p 186

Weiss-Penzias PS, Gay DA, Brigham ME, Parsons MT, Gustin MS, ter Schure A (2016) Trends in mercury wet deposition and mercury air concentrations across the US and Canada. Sci Total Environ 568:546–556

Wolfe MF, Atkeson T, Bowerman W, Burger J, Evers DC, Murray MW, Zillioux E (2007) Wildlife indicators. In: Harris R, Murray MW, Saltman T, Mason R, Krabbenhoft DP, Reash R (eds) Ecosystem responses to mercury contamination: indicators of change. CRC Press, New York, NY, p 123–189

Yang Y, Yanai RD, Schoch N, Buxton VL, Gonzales KE, Evers DC, Lampman GG (2019) Determining optimum sampling strategies for monitoring mercury and reproductive success in common loons in the Adirondacks of New York. Ecotoxicology. (this issue)

Yu X, Driscoll CT, Montesdeoca M, Evers D, Duron M, Williams K, Schoch N, Kamman NC (2011) Spatial patterns of mercury in biota of Adirondack, New York lakes. Ecotoxicology 20 (7):1543–1554

Yu X, Driscoll CT, Huang J, Holsen TM, Blackwell BD (2013) Modeling and mapping of atmospheric mercury deposition in Adirondack Park, New York. PLoS ONE 8(3):e59322

Zhang Y, Jacob DJ, Horowitz HM, Chen L, Amos HM, Krabbenhoft DP, Slemr F, Louis VL, Sunderland EM (2016) Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions. Proc Natl Acad Sci 113:526–531

Zhou H, Zhou C, Lynam MM, Dvonch JT, Barres JA, Hopke PK, Cohen M, Holsen TM (2017) Atmospheric mercury temporal trends in the northeastern United States from 1992 to 2014: are measured concentrations responding to decreasing regional emissions? Environ Sci Technol Lett 4(3):91–97

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