

Historical Trends of Mercury Deposition in Adirondack Lakes

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There is interest in historical patterns of total mercury (Hg_T) deposition because of concern over contamination to remote lakes by atmospheric Hg deposition. Sediment cores were collected from eight remote lakes in the Adirondack region of New York. Concentration and flux profiles were reconstructed from preindustrial to modern values based on ^{210}Pb dating. All cores exhibited an increase in Hg_T since 1850. The ratio of modern Hg_T flux to preindustrial Hg_T flux in sediments ranged from 1.6 to 5.7, with an average value of 3.5. The lake cores showed an increase in sediment Hg_T flux with increasing ratio of watershed area to lake surface area. Our analysis suggests that in 1850 Adirondack watersheds retained 95% of atmospheric deposition of Hg. This fraction has decreased since about 1930 to 78% currently, a value that agrees with watershed mass balance studies. Extrapolation of the relationship between sediment Hg_T flux and watershed area to lake surface area to a value of 1 suggests that the flux of atmospheric Hg_T deposition was $5.0 \mu g m^{-2} yr^{-1}$ in preindustrial times and is $8.9 \mu g m^{-2} yr^{-1}$ currently. This modern estimate of Hg deposition to lake sediments is somewhat lower (~36%) than direct estimates of atmospheric Hg_T deposition.

Introduction

Mercury (Hg) contamination in remote lakes has been largely attributed to atmospheric deposition (1, 2). The atmospheric emissions of Hg increased after 1850 with increased mining of gold, coal combustion, and other industrial activities (3). Mercury was once widely used in electrical devices, laboratory instruments, pesticides, chlorine production, batteries, paints, hat felts, gilding, manufacture of mirrors, and dental amalgams. In recent decades these uses have curtailed, and anthropogenic emissions of Hg to the atmosphere are currently largely from coal combustion and waste incineration (4, 5). Although industrial Hg consumption has decreased (6), Hg in the atmosphere over North America is increasing by approximately 1.5% per year (7). Anthropogenic sources account directly for 44% of present-day emissions of Hg (3). Remote lakes may exhibit elevated concentrations of Hg in fish tissue since Hg is characterized by long-range atmospheric transport and may be deposited considerable distances from emission sources (7). Mercury deposited to a watershed from atmospheric deposition may be revolatilized to the atmosphere. It has also been reported that 20–40% of the Hg deposited to a watershed can be transported to interconnected lake ecosystems (8).

Total mercuric ion (Hg^{2+}) is the predominant form found in aquatic environments. Although it binds firmly to sediments (8), there are other pathways that Hg^{2+} can follow. Under anaerobic conditions Hg^{2+} can be reduced to elemental Hg^0 , which can be lost from the lake by evasion (4). It can also be methylated by bacteria (4, 5). If dimethyl mercury forms, it will readily degas (5). Demethylation of Hg also results in the formation of Hg^0 (5). Monomethyl mercury is readily assimilated by aquatic biota and accumulates in biological tissues (4, 5).

Due to an interest in the biogeochemistry of Hg in remote areas, several studies have been conducted in the Adirondack region of New York State. Driscoll et al. (9) found that concentrations of total Hg (Hg_T) were higher in Adirondack lakes than other remote regions of eastern North America. Their highest values were comparable to concentrations from lakes in urban areas. In addition, elevated concentrations of Hg were observed in fish (10). In assessing the role of atmospheric deposition of Hg in controlling fish Hg concentrations, there is interest in determining deposition of Hg prior to the Industrial Revolution and in qualifying changes that have occurred over the past 150 years.

The objective of this study was to evaluate changes in Hg deposition over a historical time scale. This was accomplished by measuring the concentrations of Hg_T in sediment cores from several Adirondack lakes. Lake sediments are a useful way to examine the historical record of Hg deposition. They provide a nearly complete record since 90% of incoming Hg is retained in sediments and postdepositional movement is very limited (8). The contribution of the watershed inputs of Hg to lake sediments was also investigated.

Site Description and Methods

The Adirondack region is an area of approximately 2.4 million ha. It is largely forested and underlain by metasedimentary rocks and granitic gneisses. Soils are largely Spodosols, and the area receives approximately 100 cm of precipitation/year (9). The area of wetlands is fairly small, typically less than 10% of the land area, with riparian wetlands and those formed by beaver impoundments the two most common types of wetlands (11). Despite the relatively small area of the landscape, wetlands have been shown to play an important role in the transport and cycling of Hg in the region (9, 10).

The lakes in this study (Figure 1) are characterized by a wide range of surface areas and watershed areas (Table 1). Most of the lakes are acidic, with low values of acid-neutralizing capacity. Wetlands make up 14–40% of the watershed for the three lakes for which data are available. The study lakes included the following: (i) seven drainage lakes that receive water from precipitation, surface runoff, seepage, and stream discharge and have a surface outlet; (ii) a single seepage lake, Little Echo Pond, with no surface inlet or outlet. Little Echo Pond is a bog seepage lake with a watershed area approximately equal to the lake surface area (12). It is presumed that Hg inputs to this site would be almost entirely derived from direct inputs of atmospheric deposition.

The sediment cores used for this project were taken during 1982–1983 as part of the Paleocological Investigations of Recent Lake Acidification (PIRLA; 13). The cores were sectioned into 0.5-cm intervals and dated using ^{210}Pb activities. The dates determined by this method agreed with other markers such as wildfire charcoal, soot from locomotives, and pollen (13). Thus, it has been concluded that the

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TABLE 1. Characteristics, ^{210}Pb Flux, and Water Chemistry Data for the Adirondack Study Lakes

lake	surface area (ha)	watershed area ^a (ha)	unsupported ^{210}Pb flux ($\text{pCi cm}^{-2} \text{yr}^{-1}$)	pH	acid neutralizing capacity ($\mu\text{equiv/L}$)
Big Moose Lake	504	9555	0.64	5.10	6
Little Echo Pond	1	1	0.08	4.26	-40
Merriam Lake	8.1	242.9	0.26	4.88	-6
West Pond	10.4	108.1	0.23	5.31	10
Bear Pond	21.9	85.6	0.13	4.94	-5
Queer Lake	54.5	154.6	0.57	5.63	7.4
Upper Wallface Pond	5.5	58.3	0.22	4.93	-2
Clear Pond	70.4	600.6	0.53	7.05	100

^a Watershed area includes lake surface area.

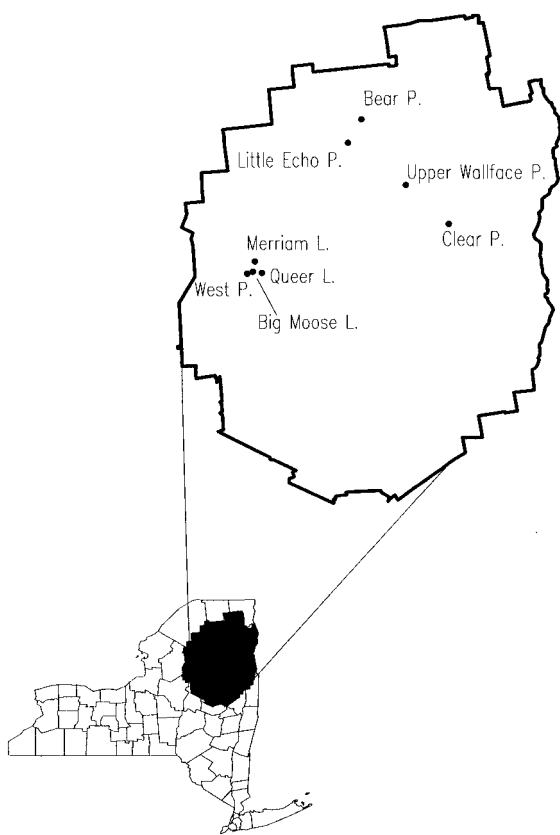


FIGURE 1. Location of the eight study lakes.

constant rate of supply model ^{210}Pb dates was accurate for PIRLA cores (13).

Total Hg concentrations in sediments were determined using an analytical method similar to that used by Engstrom et al. (8). The samples were oven-dried; digested in trace metal grade sulfuric, nitric, and hydrochloric acids; and oxidized with potassium permanganate and potassium persulfate. After prereduction with hydroxylamine hydrochloride, the samples were reduced with tin(II) chloride and analyzed for Hg_T by flameless atomic absorption. Each batch of samples included three to four samples of San Joaquin soil from the National Institute of Standards and Technology as a standard reference material.

Results and Discussion

The concentration of Hg_T in Adirondack lake sediments ranged from 0.08 to 0.20 $\mu\text{g/g}$ in the deeper portions of the core. For a given core, concentrations of Hg_T were relatively constant at lower depths. Concentrations of Hg_T increased with decreasing sediment depth at depths around 8–16 cm. The upper sediments had concentrations ranging from 0.18 to 0.50 $\mu\text{g/g}$. Five of the lakes showed a pattern of decreasing

concentrations of Hg_T in the top 2 cm. Three of these cores exhibited decreases in Hg_T concentrations of more than 10%, while the other two showed much smaller declines.

All lakes showed an increase in sediment deposition of Hg_T from 1850 values. Among drainage lakes, the preindustrial fluxes ranged from 5.1 to 11.6 $\mu\text{g m}^{-2} \text{yr}^{-1}$ while modern values ranged from 18 to 64.3 $\mu\text{g m}^{-2} \text{yr}^{-1}$ (Figure 2). Little Echo Pond, the bog seepage lake, had a preindustrial value of 3.1 $\mu\text{g m}^{-2} \text{yr}^{-1}$ and a modern value of 4.9 $\mu\text{g m}^{-2} \text{yr}^{-1}$.

Although other studies (1, 6) have shown increased sediment Hg_T deposition rates around 1850, for the majority of lakes in the Adirondacks this increase occurred at a later date. Sediment deposition of Hg_T in Little Echo Pond started to increase around 1850. However, in Big Moose Lake, Upper Wallface Pond, and West Pond, sediment Hg deposition started to increase around 1870 while the remaining four lakes began to show an increase in sediment Hg_T deposition around 1900. Although spatial differences in precipitation and atmospheric Hg deposition may have influenced the temporal pattern of sediment Hg_T deposition, drainage lakes with a larger watershed area to lake surface area ratio exhibited a delay in the response to increases in atmospheric emissions and deposition of Hg_T that may be indicative of watershed retention of Hg_T . While three sediment cores showed an expected decline of Hg_T deposition in recent sediments (~1950–1970) presumably in response to decreases in Hg emissions, five sediment cores did not exhibit this pattern. The bog seepage lake, Little Echo Pond, showed the greatest decrease (~44%) in Hg_T deposition in recent sediments.

Profiles of ^{210}Pb and values for unsupported ^{210}Pb flux of these cores are given in Binford et al. (13). The global average ^{210}Pb flux is 0.5 $\text{pCi cm}^{-2} \text{yr}^{-2}$. Values above this would be an indication of focusing, while values below this would show an under-accumulation. The values for our cores ranged from 0.08 to 0.61 $\text{pCi cm}^{-2} \text{yr}^{-2}$ (Table 1; 13). There were no values that were significantly higher than 0.5 $\text{pCi cm}^{-2} \text{yr}^{-2}$, which would suggest that focusing was not a major problem in these cores. The lowest value (0.08 $\text{pCi cm}^{-2} \text{yr}^{-2}$), which indicates a large under-accumulation, occurred in Little Echo Pond. This pattern may partially help explain the relatively low values of Hg_T flux (modern and preindustrial) in sediments of Little Echo Pond (discussed below).

The flux ratios were determined as the ratio of modern to preindustrial Hg_T deposition rates (Table 2). The modern values are the most recent sediments for each lake (~1982), while the preindustrial values are averages of several values for sediments deposited in the mid-1800s prior to the increase in Hg deposition. The average flux ratio for the eight Adirondack lakes was 3.5, which is in good agreement with the average of 3.7 reported by Swain et al. (1) for seven lakes in Minnesota and Wisconsin (Table 2). The values were also similar to other Canadian and European studies but were higher than flux ratios reported for some arctic lakes (Table 2; 14).

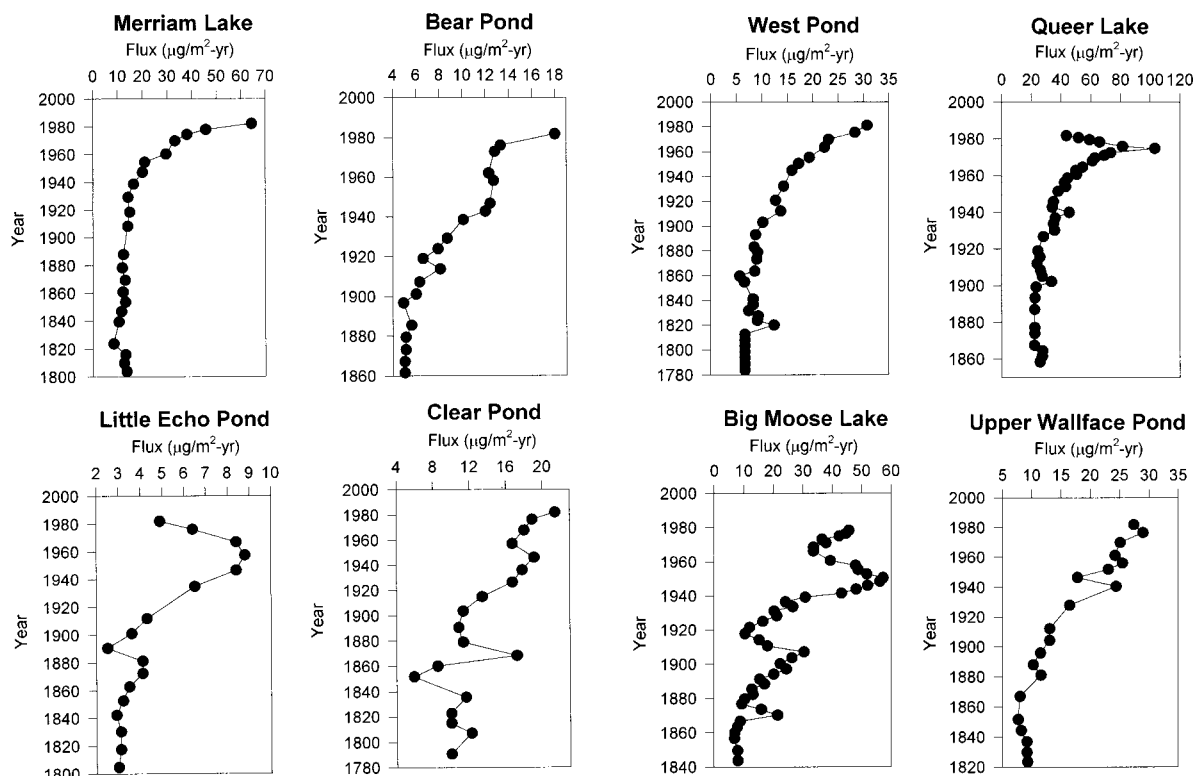


FIGURE 2. Total mercury deposition rates by year in sediment cores dated by ^{210}Pb in study lakes.

TABLE 2. Lake Flux Values and Flux Ratios of the Deposition of Total Mercury (Hg_T) in the Sediments of Adirondack Lakes^a

lake	modern flux ($\mu\text{g m}^{-2} \text{ yr}^{-1}$) and year used	preindustrial flux ($\mu\text{g m}^{-2} \text{ yr}^{-1}$) and years used	flux ratio
Big Moose	42.5 (1975)	7.5 (av 1840–1863)	5.7
Little Echo	4.9 (1982)	3.1 (av 1800–1850)	1.6
Merriam	64.3 (1982)	11.6 (av 1800–1850)	5.5
West	30.7 (1981)	6.7 (av 1785–1820)	4.6
Bear	18.0 (1982)	5.1 (av 1860–1900)	3.5
Queer	38.6 (1982)	23.8 (av 1860–1900)	1.6
Upper Wallface	27.4 (1982)	8.7 (av 1820–1850)	3.2
Clear	21.4 (1982)	10.0 (av 1800–1850)	2.1
average			3.5
average of Swain et al. (7)		midcontinental U.S.	3.7
summary of values compiled by Landers et al. (14)		Finland	4.4
		Sweden	3.0
		W. Canada	2.7
		Quebec	2.1
		Russia	1.2
		Alaska	1.1

^a These rates are compared with other studies in the literature.

If Hg is transported from the watershed to a lake, then we would anticipate the amount of sediment Hg_T deposition to increase with increasing watershed to lake surface area. All Adirondack lakes showed a good relationship between sediment Hg_T deposition and the ratio of watershed area to lake surface area, except for Queer Lake (Figure 3). The reason for this outlier is unknown. Queer Lake has two outlets; one that is important under high flow conditions, and the other that is important under low flow conditions. Additionally, Queer Lake was the only site where ^{210}Pb dates are regarded as unreliable (13). As a result of the discrepancy, the values of sediment Hg_T deposition for Queer Lake were not included in the regression analysis. The sediment Hg_T deposition for

Little Echo Pond fell somewhat below the regression analysis as a function of the ratio of watershed area to lake surface area. This pattern may have been due to the under-accumulation of ^{210}Pb observed for this site.

Extrapolation of the regression lines to an area ratio of 1 is thought to represent a watershed with only direct Hg_T deposition to the lake surface (watershed area = lake surface area). For the modern values of sediment Hg_T deposition, a watershed area to lake surface area of 1 results in a value of $8.9 \pm 2.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$. This flux is somewhat lower than the value of $12.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$ found by Engstrom et al. (8) for lakes in Minnesota and Wisconsin but is generally consistent with the range of $10\text{--}15 \mu\text{g m}^{-2} \text{ yr}^{-1}$ reported for several studies in midcontinental North America (1). Wet deposition of Hg_T was found to contribute $9.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in the Lake Champlain basin and ranged from 5.7 to $11.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$ (average = $8.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$) for three sites in Michigan (15). Assuming that dry deposition of Hg is approximately 50% of wet deposition (16), our estimate of sediment deposition of Hg_T ($8.9 \mu\text{g m}^{-2} \text{ yr}^{-1}$) appears to be somewhat lower than the total atmospheric deposition of Hg_T to the region. One possibility for the difference between our sediment analysis and studies of atmospheric Hg deposition is an increase in atmospheric deposition of Hg_T from the time when the sediment cores were collected (1982). If it is assumed that the airshed of the Adirondacks follows a national trend of decreasing Hg emissions (6), then this does not seem to be a reasonable explanation for the discrepancy in Hg_T deposition. Alternative explanations may be that (i) dry deposition is a lower fraction of wet deposition than previously thought and/or dry deposition to lake surfaces may be lower than to the surrounding forest watershed, (ii) significant Hg_T is lost from the lake outlet, and (iii) Hg_T^{2+} is reduced to Hg^0 and subsequently lost by evasion before sedimentation. This reductive pathway has been shown to be significant in the cycling of Hg^0 (16). A mass balance study of a seepage lake in Wisconsin indicated that losses of Hg^0 by evasion accounted for 6.4% of the deposition of Hg_T , while other lakes studied ranged from 10% to 50% of Hg_T deposition lost

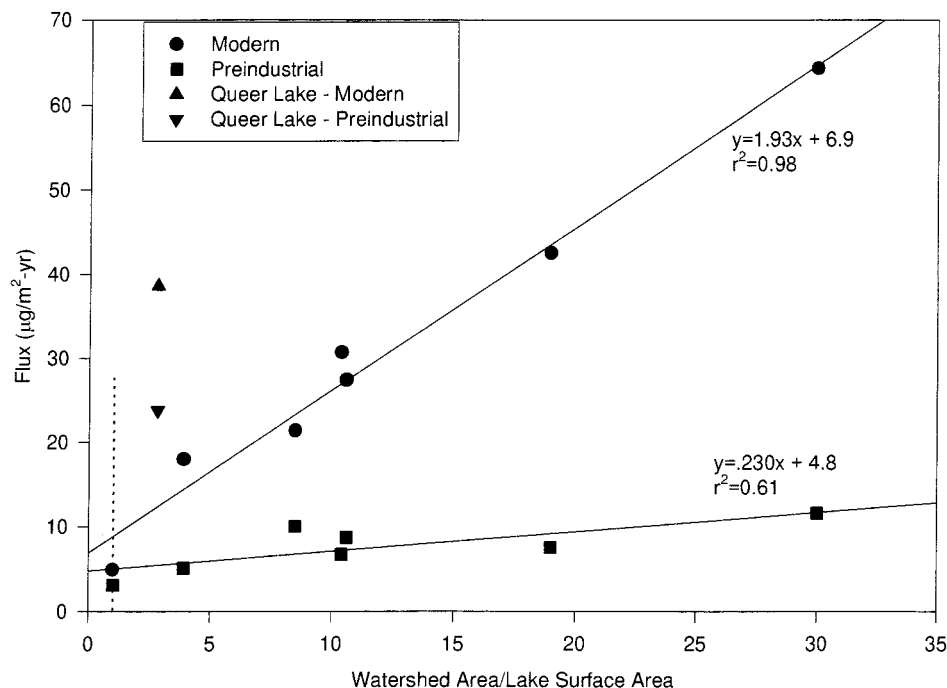


FIGURE 3. Modern and preindustrial (~1850) rates of sediment mercury deposition as a function of the ratio of the watershed area to the lake surface area. Queer Lake was observed as an outlier and not considered in the regression analysis. The regression analysis is extrapolated to a value of 1 (watershed area = lake surface area) to estimate direct deposition of mercury to the land surface.

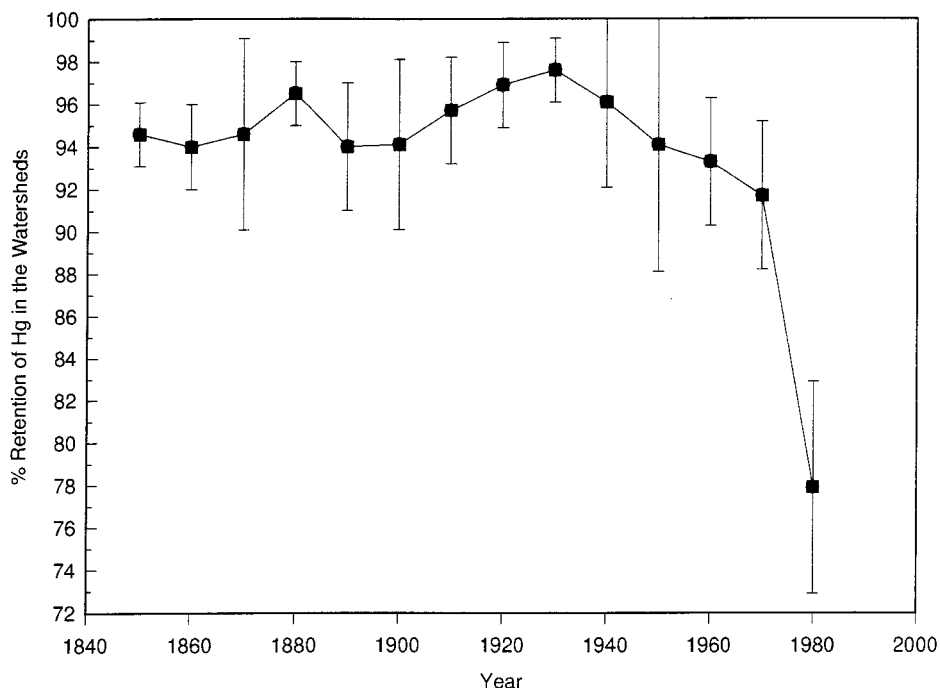


FIGURE 4. Percent retention of total mercury in the watersheds for each decade from 1850 to 1980. The error bars represent standard error in the calculations.

by evasion (17). The preindustrial value of sediment Hg deposition in the Adirondacks ($5.0 \pm 1.3 \mu\text{g m}^{-2} \text{yr}^{-1}$ from intercept of Figure 3) was slightly higher than the value of $3.7 \mu\text{g m}^{-2} \text{yr}^{-1}$ found by Engstrom et al. (8).

The slope of the relationship between sediment Hg_T deposition and the ratio of watershed area to lake surface area represents the additional flux of Hg_T beyond direct atmospheric deposition to the lake surface with each unit increase in the ratio of the surface areas. As the slope of a plot of Hg deposition as a function of watershed area to lake surface area approaches 0, the contribution of Hg to lake sediments from the watershed becomes insignificant (8). The

slopes of both the modern and preindustrial values of Hg_T deposition as a function of watershed area to lake surface area were significantly different from 0 ($p < 0.0001$ and $p = 0.0392$, respectively; Figure 3). Engstrom et al. (8) obtained values of $3.27 \mu\text{g m}^{-2} \text{yr}^{-1}$ for the modern slope and $0.83 \mu\text{g m}^{-2} \text{yr}^{-1}$ for the preindustrial slope on a plot of sediment Hg deposition as a function of watershed area to lake surface area, while our results were $1.93 \mu\text{g m}^{-2} \text{yr}^{-1}$ for modern and $0.23 \mu\text{g m}^{-2} \text{yr}^{-1}$ for preindustrial values. It appears that the watersheds of the Adirondack lakes transport less Hg_T to lakes than observed for Minnesota and Wisconsin (8). Since both areas have received similar atmospheric Hg deposition

since preindustrial times, the lower transport of Hg_T indicates that Hg deposited to the Adirondack landscape by atmospheric deposition will likely continue to be transported from the watershed to lakes for a considerable period in the future. This pattern may reflect the fact that five of the seven lakes studied by Engstrom et al. (8) were seepage lakes with two of the lakes exhibiting occasional drainage from the watershed, while our lakes are mostly (seven out of eight) drainage lakes. The increase in the slope from $0.23 \mu\text{g m}^{-2} \text{yr}^{-1}$ in preindustrial time to $1.93 \mu\text{g m}^{-2} \text{yr}^{-1}$ in modern times (Figure 3) is an indication of the importance of the watershed area to lake surface area ratio in the supply of Hg to Adirondack lakes under current conditions. The watershed contribution of Hg_T to lake sediments has increased markedly in the Adirondacks since 1850.

The percentage of Hg_T entering the watershed that is transported to the lake can be estimated from a plot of sediment Hg deposition as a function of the ratio of watershed area to lake surface area (Figure 3) by dividing the slope by the intercept. By using this approach, it is assumed that (i) extrapolation of the plot of sediment Hg deposition as a function of the ratio of watershed area to lake surface area to a value of 1 is representative of atmospheric Hg deposition and (ii) losses of Hg_T from the watershed are deposited in lake sediments. The resulting calculations showed that 78% of atmospheric Hg_T deposition was retained in the watershed for the modern period and 95% for preindustrial conditions. Engstrom et al. (8) determined modern and preindustrial values of watershed Hg retention to be 74% and 78%, respectively, of atmospheric deposition. The 17% change in retention of Hg_T that we found for the Adirondacks likely reflects changes in the watershed supply of Hg_T to lakes over the last 150 years. The watershed retention of atmospheric Hg_T deposition for each decade from 1850 to 1980 was calculated for the lakes in this study (Figure 4). This analysis showed that watershed Hg_T retention was high and did not change from 1850 to 1930 (mean value 95.3%; $p > 0.05$). Values decreased slightly but significantly ($p < 0.05$) from 1930 to 1970 and decreased markedly for 1970–1980 ($p < 0.05$). Our modern value of watershed Hg_T retention (78%) is in good agreement with other watershed Hg mass balance studies. Values for watershed Hg_T retention of atmospheric deposition of 74% (8) and 76% (11) have been determined for watershed studies in the upper Midwest and the Adirondacks, respectively. Although watershed retention of Hg_T is directly related to the size of the watershed, the amount of wetlands in the watershed must also be considered. St. Louis et al. (18) found an average Hg retention of 59.4% for five catchments containing a large quantity of wetlands in Ontario. Since the watershed retention of Hg_T decreases when wetlands are present (10, 18, 19), an increase in wetland area may possibly explain the decline in watershed Hg_T retention since the 1930s in the Adirondacks.

The observation that 22% of the Hg_T entering the watershed from atmospheric deposition is transported to the lakes as opposed to 5% in preindustrial times may have important management implications for the region. While factors such as acidic deposition and acidification of drainage water could have contributed to this pattern, another possible mechanism may be the development of wetlands associated with the activity of beavers (11, 20). Due to trapping and habitat loss in northern New York, there were less than 1000 beavers by 1820 and as few as 5–10 estimated in 1895 (21). Beavers were released in New York from 1901 to 1907 and have responded well. Current estimates of beavers in the

Adirondack Park are 14 000 to 18 000 animals (21). Beaver impoundments help to create riparian wetlands (20). In addition, riparian wetlands and those created by beaver impoundments are the two most common types in the Adirondacks (11). The development of wetlands by beaver activity may facilitate the transport of Hg_T from atmospheric deposition and soil pools in watershed areas that were previously uplands to downstream lake ecosystems.

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