Spatial Trends and Historical Deposition of Mercury in Eastern and Northern Canada Inferred from Lake Sediment Cores

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Recent and historical deposition of mercury (Hg) was examined over a broad geographic area from southwestern Northwest Territories to Labrador and from the U.S. Northeast to northern Ellesmere Island using dated sediment cores from 50 lakes (18 in midlatitudes (41−50°N), 14 subarctic (51−64°N) and 18 in the Arctic (65−83°N)). Distinct increases of Hg over time were observed in 76% of Arctic, 86% of subarctic and 100% of midlatitude cores. Subsurface maxima in Hg depositional fluxes (μg m^{-2} y^{-1}) were observed in only 28% of midlatitude lakes and 18% of arctic lakes, indicating little recent reduction of inputs. Anthropogenic Hg fluxes adjusted for sediment focusing and changes in sedimentation rates ($\Delta F_{adj,F}$) ranged from −22.9 to 61 μg m^{-2} y^{-1} and were negatively correlated ($r = -0.57$, $P < 0.001$) with latitude. Hg flux ratios (FRs; post-1990)/pre-1850) ranged from 0.5 to 7.7. The latitudinal trend for Hg $\Delta F_{adj,F}$ values showed excellent agreement with predictions of the global mercury model, GRAHM for the geographic location of each lake ($r = 0.933$, $P < 0.001$). The results are consistent with a scenario of slow atmospheric oxidation of mercury, and slow deposition of reactive mercury emissions, declining with increasing latitude away from emission sources in the midlatitudes, and support the view that there are significant anthropogenic Hg inputs in the Arctic.

Introduction

Elevated mercury (Hg) in fish has been well documented in midlatitude North American lakes (*1, 2*), and Hg has emerged as a priority contaminant in lakes and rivers of Arctic and subarctic North America (*3*). Hg concentrations in fish are ultimately linked to bioaccumulation of methyl Hg in lakes and their catchments (*4*). Recent studies using Hg isotopes have shown that methyl Hg concentrations in fish responded rapidly to changes in (inorganic) Hg deposition in a lake in the Experimental Lakes Area in northwestern Ontario with the increase coming almost entirely from deposition to the lake surface (*5*). Hg deposition has been modeled using atmospheric transport and deposition models (*6, 7*) which predict significant anthropogenic Hg deposition in Arctic and subarctic regions although declining with increasing north latitude. Yet, there is only limited validation of model predictions because routine Hg wet deposition measurements are limited to south of about 50°N (*8*).

Lake sediments have been shown to be reliable archives for estimating historical Hg accumulation (*9, 10*) although it has been noted that sediments are not especially responsive to changes in Hg deposition over the past 10−20 years owing to slow Hg transport through catchment soils (*11*). Lake sediments have been used to estimate current and historical Hg deposition in numerous midlatitude lakes in North America (*12*−*16*). Recent studies by Mills et al. (*16*) on lakes in south-central Ontario lakes and by Engstrom et al. (*15*) on Minnesota lakes provide anthropogenic Hg enrichment and deposition data for over 200 midlatitude (44−48 °N) lakes. Reviews of Hg deposition to Arctic and subarctic lakes based on sediment cores of early to late-1990s data noted that there was a lack of information on Hg deposition in the eastern Arctic and northern Québec, where no cores from lakes north of 55°N latitude had been analyzed (*17, 18*). Recently Fitzgerald et al. (*19*) studied Hg deposition in five lakes in the north slope of Alaska, and Outridge et al. (*20, 21*) reported on historical sedimentary profiles of Hg in two lakes in the central Canadian Arctic archipelago. Overall, however, little information is available on deposition of Hg to subarctic and Arctic lakes in North America compared to midlatitude regions. Thus while knowledge of Hg deposition in lake sediment cores over large spatial scales could aid understanding of spatial and temporal trends of Hg in fish, the current geographical coverage is poor.

Our objectives were to quantify latitudinal and longitudinal trends of anthropogenic Hg deposition in eastern and northern North America, to investigate variations in Hg deposition, to examine relationships with lake area, catchment/lake area ratio and sedimentation rates, and to compare our results with model predictions.

Materials and Methods

Study Design and Site Selection. Sediment cores were obtained during the period 1998−2005 from lakes in the Canadian Arctic archipelago, subarctic lakes in the southwestern Northwest Territories, Manitoba, Ontario, Québec, and Labrador, and midlatitude lakes in Ontario, Québec, and northeastern U.S. (Figure 1). Full details on the lake characteristics are provided in Supporting Information (SI) Table S1. Lakes were assigned to Arctic (>65°N and above the treeline), subarctic (Labrador, Northern Québec, Ontario

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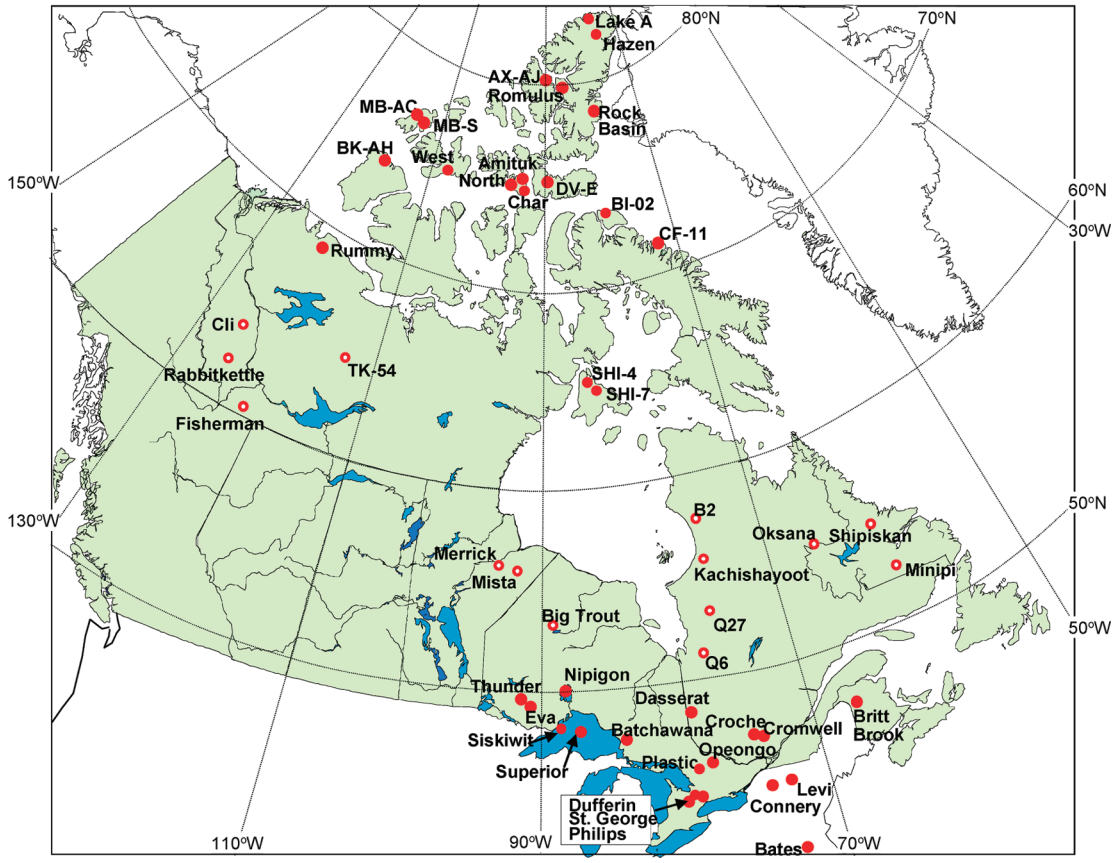

FIGURE 1. Location of the 50 study lakes. Midlatitude lakes were south of 51°N, subarctic lakes (open symbols) from 51−65°N and Arctic lakes north of 65°N. See SI Table S1 for additional information on each lake.

TABLE 1. Pearson Correlation Coefficients[a] for Correlations of Lake Characteristics with Latitude and Longitude

	N	latitude	long-itude	precip[b] (mm)	% organic carbon	log A_L	log A_C	A_C/A_L	focusing factor[c]	Al flux ratio[d]
precipitation (mm)	50	−0.941**	−0.519**							
% organic carbon	47	−0.421*	−0.278	0.424*						
Log A_L	50	−0.224	−0.077	0.166	−0.150					
Log A_C	50	−0.125	−0.107	0.092	−0.241	0.897**				
A_C/A_L	50	0.138	−0.114	−0.112	−0.191	−0.093	0.241			
focusing factor	49	0.456**	0.120	−0.429**	−0.357*	0.092	0.212	0.444**		
Al flux ratio	48	−0.139	−0.155	0.142	−0.009	−0.056	−0.013	0.014	0.054	
sedimentation rate	49	0.347*	0.172	−0.338*	−0.368*	0.017	0.147	0.356	0.717**	0.185
sedimentation ratio[e]	49	−0.114	−0.116	0.105	−0.007	−0.056	−0.054	−0.004	0.014	0.915**

[a] ** and * = Significant Pearson correlation coefficients at $P = 0.01$ and 0.05, respectively, using uncorrected probabilities. [b] Average (1970−2000) precipitation at nearest Environment Canada (http://www.climate.weatheroffice.ec.gc.ca/climateData/canada_e.html) or NOAA (http://cdo.ncdc.noaa.gov/cgi-bin/climatenormals/climatenormals.pl) station. [c] Focusing factor based on average excess ^{210}Pb divided by known or estimated ^{210}Pb for the latitude of the individual lake. For further details on estimated ^{210}Pb see the Supporting Information. [d] Aluminum (Al) flux = ((recent (post-1990)/pre-1850) were calculated to estimate erosional/aoelian inputs. [e] Sedimentation ratio = CRS estimated sediment flux, g m^{-2} y^{-1} ((recent (post-1990)/pre-1850).

and Manitoba >51°N and Northwest Territories), and midlatitude (south of 51°N) groups.

Sample Collection and Dating. In brief, cores (6−10 cm diam) were obtained from the deepest point in each lake avoiding steep sloped areas. Cores were extruded and sliced into 0.5 or 1 cm sections (SI Table 1). Sediment cores were dated using the ^{210}Pb and/or ^{137}Cs methods, and sedimentation rates and dates were estimated using the constant rate of supply (CRS) model (*22*). Only cores with interpretable ^{210}Pb or ^{137}Cs stratigraphy were selected for analysis; they typically yielded 10−20 post-1800 core sections or horizons, per core. Further details on collection, dating, and analysis are provided in the Supporting Information.

Mercury and Multielement Analysis. Freeze-dried subsamples were placed in Teflon vessels and acid-digested with a mixture of nitric and hydrochloric acids and hydrogen peroxide (ratio 9:2:1) in a high pressure microwave oven. Total Hg was determined by cold vapor atomic absorption spectrometry. A subset of cores were analyzed by direct combustion using a DMA-80 (Milestone Instruments, Shelton, CT). Total Al and Zn were determined by inductively coupled plasma-mass spectrometry (ICP-MS) (PQ-2, VG Elemental) and manganese (Mn) and iron (Fe) using ICP-atomic emission spectrometry (AES). Organic carbon (OC) was determined by CHN analyzer in subsamples from 47 of 50 cores.

Flux Calculations and Statistical Analysis. Recent and preindustrial horizons in each core were defined by the median ages assigned to each core section using the CRS

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model. Hg fluxes, flux ratios (FR), and enrichment factors (EFs) were calculated as

Flux (F) (μg m^{-2} y^{-1}) = [dry wt concn] × ^{210}Pb-derived sedimentation rates for each core horizon.

Anthropogenic flux, ΔF (μg m^{-2} y^{-1}) = F_{recent} (post-1990) − F_{preind} (preindustrial; pre-1850), FR = F_{recent}/F_{preind} and EF = recent (post-1990)/preindustrial Hg concentrations.

Fluxes of Al were calculated to investigate inputs of a primarily lithogenic element and Al flux ratios [(recent (post-1990) − pre-1850)/pre-1850] were calculated to estimate erosional and aeolian inputs. After confirming a strong correlation of Al flux ratio and the ratio of ^{210}Pb derived recent and preindustrial sedimentation rates ($r = 0.915$, $P < 0.001$, $N = 34$), Hg fluxes for each horizon were adjusted for sedimentation and an anthropogenic Hg flux ΔF_{adj} was calculated using the equation of Perry et al. (*13*):

$$\Delta F_{adj} = F_{recent} - F_{pre\text{-}ind} - (F_{pre\text{-}ind} \times \text{sedimentation ratio} - F_{pre\text{-}ind}) \quad (1)$$

As noted by Perry et al. (*13*) if the pre-1850 sedimentation rate equals the post-1850 sedimentation rate, the sedimentation ratio = 1 and no adjustment is made. However, in horizons where the sedimentation rate is higher, the ratio adjusts for the dilution effect of increased sedimentation.

Sediment particle focusing factors (FF) were estimated for all cores dated by dividing the observed ^{210}Pb flux (Bq m^{-2}y^{-1}) by the predicted ^{210}Pb flux for the same latitude based on soil ^{210}Pb measurements from 41.5 to 81.5°N latitude (see SI Table S2 and Figure S1 and references therein). Recent fluxes and $\Delta F_{adj,F}$ were adjusted by dividing by FF.

Correlations were performed using Microsoft Excel and Systat 12 (Systat Software Inc. Chicago IL). Lake area (A_L), catchment area (A_C) and Hg fluxes were log transformed to yield normally distributed data (Shapiro−Wilk test; $P > 0.10$).

Modeled Hg Deposition. The global/regional atmospheric heavy metals model (GRAHM) (*6, 23*) was used to predict terrestrial anthropogenic Hg fluxes for each lake location. A description of this model is given by Dastoor and Laroque (*6*) and it was recently modified to include parametrizations for halogen-mediated mercury chemistry, deposition, and re-emission of mercury during spring and summer in the Arctic (*23*). Model simulations were performed for year 2001 at 50 km horizontal resolution using anthropogenic Hg emissions for year 2000 and terrestrial and oceanic emissions as described in Dastoor et al. (*23*). Net deposition of Hg to the surface in the geographic region of each of the 50 lakes was predicted based on their latitude/longitude. In Arctic and subarctic regions the model simulates rapid revolatilization of mercury in addition to the deposition represented by mercury depletion events (MDEs), which are subtracted from the deposition.

Results

Lake Characteristics, Sedimentation Rates, and Elemental Profiles. The lakes ranged widely in surface area (MB-AC = 0.0017 km^2; Superior = 82170 km^2); 37 of 50 were <10 km^2 (SI Table S1). Lake and catchment areas, as well as A_C/A_L were unrelated to lake latitude or longitude (Table 1). Most cores exhibited near-exponential declines in excess^{210}Pb with depth, indicating relatively constant rates of sediment accumulation (see SI Figure S2). Sedimentation rates based on the CRS model ranged from 10 to 1000 g m^{-2} y^{-1} (SI Table S1) and were not significantly correlated with latitude, A_L or A_C (Table 1). Sedimentation rates were generally low and the top 10 cm of most cores included sediments deposited over the past ~150 years. Sedimentation rate was significantly correlated with focusing factor ($r = 0.700$; $P < 0.001$). Sedimentation rates were >10% higher in the period 1990−2000 than in the years 1800−1900 in 27 of 49 lakes; 11 of 49 had 2× higher rates. The increased sedimentation estimated from the ratio of the CRS sedimentation rates in recent and preindustrial horizons, was unrelated to latitude, longitude, average sedimentation rate, or to average annual precipitation for each location (Table 1).

Concentrations of Al, an indicator of geogenic inputs, were available for 35 lakes and showed relatively little variation with deposition year with only 12 lakes having positive correlations and 2 having negative (SI Table S3). Al concentrations within lakes were generally not correlated with sedimentation rate in Arctic and subarctic lakes (SI Table S3). Zn concentrations were correlated with sedimentation rate in <20% of Arctic and subarctic lakes (SI Table S3). However, a slightly higher frequency of positive correlations of both Al and Zn with sedimentation rates was found in midlatitude lakes (22 and 36%, respectively). Overall, sedimentation rate did not appear to have a large lithogenic component, i.e., from erosion or aeolian inputs.

The higher recent sedimentation rates estimated using the CRS model could also be due to the flattening of the slope of the ^{210}Pb activity profile near the sediment surface due to bioturbation or to diagenetic dilution of the ^{210}Pb due to accumulation of Fe oxides at the surface (*19, 24*). Bioturbation is unlikely to be a major factor because almost all these lakes are all cold, oligotrophic systems (*25*), however, no quantitative assessment of density of benthic fauna of each lake was available. As for Fe diagenesis resulting in increased Fe in surface sediments and thus increased dry mass, total Fe concentrations increased with deposition year in 10 of 18 (56%) midlatitude cores but in only 2 of 17 arctic lakes (SI Table S3). Thus Fe diagenesis resulting in increased Fe in surface sediments (and thus increased dry mass in surface horizons) may have played a role in influencing estimated sedimentation rates at least in midlatitude lakes as discussed by Gubala et al. (*24*).

Our results for sedimentation rates in Arctic lakes are in general agreement with several recent studies which have shown higher sedimentation in late 20th century horizons compared to 19th century horizons (*19, 20*). Lindeberg et al. (*26*) concluded that changes in the influx of material from regional aeolian activity resulted in large fluctuations in Hg and Pb concentrations in pre-19th century sediments of Greenland lakes. On the other hand, Lockhart et al. (*27*) did not observe differences in recent and preindustrial sedimentation rates in a series of 16 cores ranging from central midlatitude Canada to Lake Hazen. Mean annual snowfall has increased in the central Canadian archipelago (*28*) suggesting greater snowmelt runoff leading to greater erosional inputs in some catchments. The disappearance of shallow ponds in the islands of the Canadian Arctic archipelago has been attributed to increased evaporation/precipitation ratios (*29*) may be an indication of the more rapid drying of lake catchments which could result in greater aeolian inputs for some lakes.

Percent OC of surface sediments declined weakly with latitude (Table 1) ($r = -0.421$, $P = 0.003$), whereas sediment particle focusing factor increased with latitude ($r = 0.456$, $P = 0.005$). Sediment OC has often been used to normalize Hg results because it is assumed that most catchment related Hg enters lakes associated with organic matter. Indeed % OC was positively correlated with Hg concentrations in 15 of 47 lakes and % OC increased with year of deposition in the same lakes (SI Table S3). Bindler et al. (*30*) also found the % OC also increased over time in the three dated cores from Greenland. Outridge et al. (*20, 21*) reported increasing % OC in Amituk Lake, as well as in lake DV09 on Devon Island, and also noted a strong correlation of Hg concentrations with algal-derived organic matter. We found that % OC was >10% higher in the surface section compared with horizons dated to approximately 1800−1900 in 26 of 47 cores for which we

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TABLE 2. Mean Concentrations of Hg in Surface Samples, Percent of Lakes with Subsurface Maxima, Mean Flux Ratios (FR), Recent and Pre-Industrial Fluxes, and Adjusted Anthropogenic Fluxes ($\Delta F_{adj,F}$)

region	N[a]	95% CI	μg/g Hg concn	max subsurface flux[b]	max subsurface concn[b]	FR[c]	Hg F_{recent}[d]	Hg F_{preind}[d]	Hg $\Delta F_{adj,F}$[e]
Arctic	18	mean	0.08	18%	17%	2.16	11.4	6.6	2.8
(65−83°N)		95% CI	0.04			0.53	6.4	3.4	2.0
subarctic	14	mean	0.13	25%	33%	2.50	26.9	14.8	7.5
(51−64°N)		95% CI	0.03			0.75	11.8	9.8	5.9
midlatitude	18	mean	0.24	28%	22%	3.56	63.5	24.9	26.8
(41−50°N)		95% CI	0.08			1.00	16.9	9.9	7.2
overall	50	mean	0.15	24%	24%	2.76	34.5	15.7	12.5
		95% CI	0.04			0.48	9.7	5.1	4.4

[a] Arithmetic mean Hg concentrations (ug/g dry wt) are reported for 50 lakes. Fluxes are reported 49 lakes − individual results are presented in SI Table S4. [b] Percent showing maximum concentrations or fluxes in subsurface horizons based on two consecutive near surface samples having >10% lower concentrations or fluxes. [c] FR = flux ratio = F_{recent}/F_{preind}. [d] Recent and preindustrial fluxes (μg m^{-2} y^{-1}) corrected for particle focusing. [e] Adjusted anthropogenic flux (μg m^{-2} y^{-1}) = $\Delta F_{adj,F}$ = (recent flux-preindustrial flux − (preindustrial flux × SR − preindustrial flux))/FF where SR = recent to preindustrial sedimentation ratio. See eq 1.

TABLE 3. Results of Simple Correlations of $\Delta F_{adj,F}$, F_{recent}, and $F_{pre\text{-}ind}$, and Flux Ratios (FR) of Mercury with Various Lake Characteristics[a]

correlation	comparison	r	P	N
with latitude/longitude	$\Delta F_{adj,F}$ vs latitude	**−0.572**	**<0.001**	49
	F_{recent}[b] vs latitude	**−0.684**	**<0.001**	49
	F_{preind}[b] vs latitude	**−0.501**	0.001	49
	FR vs latitude	**−0.295**	0.040	49
	FR vs longitude	−0.261	0.070	49
with % organic carbon[c]	$\Delta F_{adj,F}$ vs sediment % OC	0.195	0.189	46
	F_{recent} vs % OC	**0.301**	**0.040**	46
	F_{preind} vs % OC	**0.324**	**0.027**	46
with lake properties (all lakes)	$\Delta F_{adj,F}$ vs log A_L	0.000	0.998	49
	$\Delta F_{adj,F}$ vs A_C/A_L	−0.010	0.945	49
with midlatitude lake catchment area[d]	F_{recent} vs A_C	**−0.569**	0.013	18
	F_{preind} vs A_C	**−0.562**	0.015	18

[a] $\Delta F_{adj,F}$, recent, and preindustrial fluxes were log transformed for all correlations. Statistically significant relationships are bolded. [b] Recent and preindustrial fluxes (μg m^{-2} y^{-1}) corrected for particle focusing. [c] %OC = % organic carbon of recent (post 1990) horizons. [d] For midlatitude lakes only.

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had OC profiles while only 3 of 47 showed a >10% decrease. The increased %OC could be due to progressive loss of carbon following burial as shown by Gälman et al. (*31*) for a varved lake sediment in northern Sweden. Alternatively, increased autochthonus production could be occurring, particularly in Arctic lakes (*19, 20*). Given that % OC was significantly correlated with latitude while the sedimentation ratio was not (Table 1), we concluded that adjustment of fluxes using the sedimentation ratio (eq 1) was more appropriate than % OC since the latter might introduce a bias for evaluating latitudinal trends.

Recent and Preindustrial Hg Fluxes. Anthropogenic mercury flux, $\Delta F_{adj,F}$, ranged from −22.9 to 61 μg m^{-2} y^{-1} averaging 2.8, 7.5, and 26.8 μg m^{-2} y^{-1} in Arctic, subarctic, and midlatitude lakes, respectively (Table 2). Hg $\Delta F_{adj,F}$ had a significant negative correlation with latitude ($r = -0.572$; $P = <0.001$) (Table 3), but was not correlated with lake area, A_C/A_L or % OC in sediment. The lowest value $\Delta F_{adj,F}$ was for Rabbitkettle Lake in southwestern NWT (SI Table S4). Omitting this lake, which had higher Hg fluxes than other subarctic lakes despite its remote location, improved the correlation with latitude (Figure 2A, $r = -0.670$; $P < 0.001$).

The net anthropogenic Hg deposition fluxes simulated by GRAHM for the region of each lake declined significantly with latitude (Figure 2B, $r = 0.933$, $P < 0.001$). Individual predicted values are given in SI Table S4. These predicted values represent the average deposition within the model grid cell for a single year (2001).

Hg F_{recent} (corrected for particle focusing but not for sedimentation) also showed significant negative correlation with latitude ($r = -0.684$; $P < 0.001$; Table 3 and Figure 2C) and focus corrected preindustrial fluxes (F_{preind}) were also correlated ($r = -0.501$, $P = 0.001$; Table 3 and Figure 2D). Uncorrected recent fluxes were also significantly correlated with latitude while preindustrial fluxes showed no correlation (SI Figure S3). Results for individual lakes are provided in SI Table S4. F_{recent} and F_{preind} Hg were weakly correlated with % OC ($P = 0.040$ and 0.027, respectively) but not with longitude (data not shown). F_{recent} and F_{preind} were also correlated with precipitation ($P < 0.001$); however, precipitation and latitude were highly correlated (Table 1).

Recent Hg fluxes in midlatitude lakes have been shown to be correlated with A_C/A_Lwhile preindustrial fluxes were generally not correlated (*12, 32−34*). In this study F_{recent} and F_{preind} for Hg were not correlated with A_C/A_L, A_L, or A_C, when all lakes were combined (Table 3). However, in midlatitude lakes F_{recent} and F_{preind} for Hg were negatively correlated with A_C ($P = 0.013$ and 0.015, respectively; $N = 18$) (Table 3) (SI Figure S4). Grigal (*35*) noted that the flux of Hg via catchment streamflow showed a clear tendency for lower annual flux with increasing watershed size. Our results are thus in general agreement with the earlier studies and they

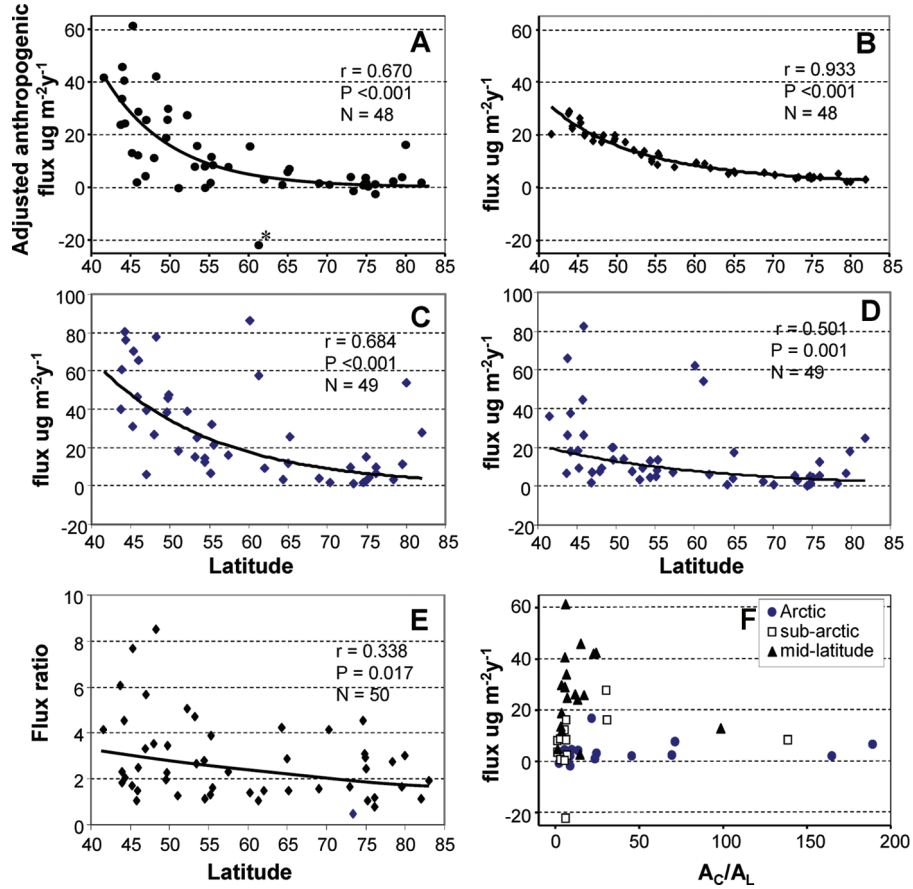

FIGURE 2. (A) Relationship of adjusted anthropogenic flux $\Delta F_{adj,F}$ for mercury vs latitude in dated sediment cores from midlatitude, subarctic and Arctic lakes, (B) comparison with net flux predicted for each lake location using GRAHM, (C) focus corrected recent fluxes, (D) focus corrected preindustrial fluxes, (E) the latitudinal trend of mercury flux ratios. All statistics for flux vs latitude are based on log transformed flux data. A single data point in A marked with a * was omitted to calculate the correlation coefficients. No significant trends of $\Delta F_{adj,F}$ with catchment to lake area ratio (A_C/A_L) were found (F).

underline the importance of catchment inputs for Hg transport, with increasing in importance in larger forested catchments common to all of the midlatitude lakes. The lack of correlation with A_C/A_L may be a consequence of large range of A_C/A_L (0.6−189) due to a more diverse group of lakes compared to other studies. Above A_C/A_L values of about 30, $\Delta F_{adj,F}$ values appear to be relatively constant and not related to geographic location (Figure 2F).

Hg FRs ranged from 0.5 to 7.7, averaging 2.76 (SI Table S4). FRs were very weakly correlated with latitude ($P = 0.04$ and 0.03) (Figure 2E). The FRs were not significantly correlated with longitude ($P = 0.070$), nor with % OC, A_C/A_L, or A_L (data not shown).

Historical Profiles of Hg. There was a general increase of Hg concentrations with deposition year in almost all cores and significant ($P < 0.05$) positive correlations of Hg concentration and year in 76% of Arctic, 86% of subarctic, and 100% of midlatitude lakes (SI Table S3). However, 18% of arctic lakes, 33% of subarctic lakes, and 22% of midlatitude lakes had subsurface maxima for Hg concentrations and 18, 25, and 28%, respectively, when based on flux (Table 2). Plots of Hg concentrations and fluxes versus CRS dates for individual sediment cores are shown in SI Figure S5.

Overall historical trends in focus and sedimentation corrected anthropogenic Hg deposition rates (i.e., $\Delta F_{adj,F}$ for each sediment horizon analyzed) in Arctic, subarctic and midlatitude lakes are presented in Figure 3. The curves were generated by calculating the average flux for 20 year time intervals from 1840 to 1900, 10 year time intervals from 1901 to 1990, and 5 year intervals from 1991 to 2005. Results for individual lakes are provided in SI Figure S6. Generally

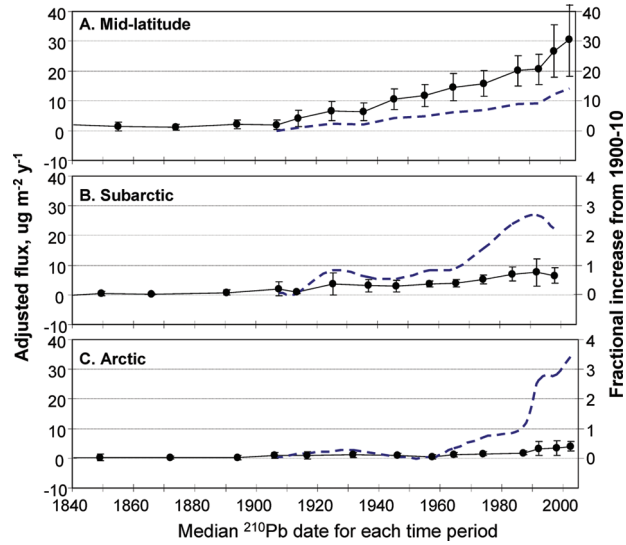

FIGURE 3. Average (±95% confidence limits) historical profiles of anthropogenic mercury deposition fluxes ($\Delta F_{adj,F}$, μg m^{-2} y^{-1}) in midlatitude, subarctic, and Arctic sediment cores for all Arctic, subarctic, and midlatitude lakes over 5−20 year time intervals. Dashed line shows the fractional increase of $\Delta F_{adj,F}$ over the same time intervals relative to values for 1900−1910. Individual lake results are given in SI Figure S5.

$\Delta F_{adj,F}$ showed a distinct increase in post-1900 horizons in midlatitude cores, post-1920−1940 in subarctic cores and post-1950 in Arctic cores. The fractional increase of $\Delta F_{adj,F}$

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relative to the anthropogenic flux in 1900−1910, shown by the dashed line in Figure 3, shows a 14.5-fold increase during the 20th century in midlatitude lakes, a 2.7-fold in subarctic, and 3.5 fold in the Arctic lakes. There is a proportionally small anthropogenic enhancement in Arctic and subarctic lakes compared to midlatitude lakes owing to relatively high background (preindustrial) Hg fluxes across all latitudes (Figure 2D; SI Figure S3).

Comparison with Other Studies of Hg Flux Ratios and Adjusted Fluxes. The range of FR values for Hg observed in this study is similar to previous reports for FRs and enrichment factors (EFs; Hg $\text{concn}_{\text{recent}}$/Hg $\text{concn}_{\text{preind}}$) in Arctic and north temperate lakes in Canada and Alaska (*17, 36*) but lower than found in the northeastern U.S. (*12*). Hg EFs for 202 lakes in south-central Ontario ranged from 0.5 to 5.1. Hg EFs for 21 lakes in West Greenland averaged 2.9 (range 0.8−11.2) (*30*), which overall, is higher than the FRs in the present study.

The Hg fluxes in North America reported by Landers et al. (*17*), based on recent horizons of cores collected in the 1980s and early 1990s over a similar range of latitude as this study, ranged from about 3 to 52 $\mu g\ m^{-2}\ y^{-1}$, whereas preindustrial fluxes ranged from 2.5 to 54 $\mu g\ m^{-2}\ y^{-1}$. These fluxes are within the range observed in this study (focus corrected). Perry et al. (*13*) reported corrected recent Hg deposition rates ($\Delta F_{adj,F}$) of 10−60 $\mu g\ m^{-2}\ y^{-1}$, averaging 25 $\mu g\ m^{-2}\ y^{-1}$ for 29 lakes in the northeastern U.S., with maximum values generally being reached between 1970 and 1990. We used the same adjustment and our average $\Delta F_{adj,F}$ for midlatitude lakes (26.8 $\mu g\ m^{-2}\ y^{-1}$) agrees well with the latter study. Two study lakes, Levi (VT) and Bates (CT) had $\Delta F_{adj,F}$ of 41 and 45 $\mu g\ m^{-2}\ y^{-1}$, respectively, similar to other lakes in that region (*13*).

Lockhart et al. (*27*) studied a series of lakes along a north−south transect and reported greatest anthropogenic enrichments in Hg occurred in central/southern Canada with a 2 fold increase over the past half century. Their fluxes for Lakes Amituk (3.9 $\mu g\ m^{-2}\ y^{-1}$) and Hazen (23 $\mu g\ m^{-2}\ y^{-1}$) were in good agreement with the present study (SI Table S4). Two more recent studies of Arctic lakes also found Hg fluxes within the range reported in this study. Bindler et al. (*30*) found recent fluxes of Hg in dated cores from three west Greenland lakes ranging from about 5 to 10 $\mu g\ m^{-2}\ y^{-1}$, and Fitzgerald et al. (*19*) estimated whole lake Hg fluxes of approximately 2.5−5.0 $\mu g\ m^{-2}\ y^{-1}$ for five lakes in northern Alaska after correcting for soil erosion.

Comparison with Predicted Trends in Fluxes for Hg. Reduction in overall atmospheric Hg emissions in North America and Europe of about 10 and 50%, respectively, between 1990 and 2000 have been reported, whereas Asian sources (mainly China) increased by 40% over the same time period (*37*). Thus, midlatitude North American lakes might be expected to show declining Hg deposition, and there is evidence of declining Hg fluxes in some studies (*13, 27, 38*). However, other recent studies of remote midlatitude lakes show limited or no decline (*15, 16*) which agrees with our observations for 72% of the 18 midlatitude lakes in this study and with the average decadal trends (Figure 3). The absence of a relatively sharp downturn reflects the slow response of lake sediments that has been attributed to the strong retention of Hg within watershed soils and biomass (*5, 13*).

Independent evidence for whether Hg deposition is increasing or decreasing in Arctic lakes is very limited. The Canadian Arctic archipelago, where almost all our high latitude study lakes were located, is very remote, and north of about 70° latitude, is influenced mainly by Asian and European atmospheric sources (*39, 40*). Analysis of time series for gaseous elemental Hg (GEM) concentrations over the period of 1995−2005 have shown no evidence of a decline at Alert on northern Ellesmere Island (*41*). Boutron et al. (*42*) reported a 40 year record of Hg in snow cores from the Greenland icecap which showed relatively constant concentrations in the 1980s, unlike increases observed in sediment cores in Greenland over the same period (*30*). Fitzgerald et al. (*19*) concluded that there was significant anthropogenic Hg deposition to lakes in the Toolik Lake area of northern Alaska due to a combination of inputs from polar sunrise MDEs and reactive gaseous Hg dry deposition. Using a relationship between Hg concentrations in precipitation and ^{210}Pb activity from remote northern hemispheric locations they estimated a Hg flux for northern Alaska of 2.2 ug $m^{-2}\ yr^{-1}$ which is similar to the average $\Delta F_{adj,F}$ of 2.8 $\mu g\ m^{-2}\ yr^{-1}$ for 18 Arctic lakes in this study. Brooks et al. (*43*) estimated net gains of Hg of 0.7 ± 0.2 $\mu g\ m^{-2}$ at Barrow, AK as a result of spring time MDEs.

The GRAHM predicted average (±SD) Hg fluxes of 4.1 ± 1.1, 11 ± 3.1, and 22 ± 3.9 $\mu g\ m^{-2}\ y^{-1}$ for the Arctic, subarctic, and midlatitude lake locations, respectively. This was in very good agreement with the general trend seen for $\Delta F_{adj,F}$ in Figure 2A and the average fluxes for each region (Table 2). Other Hg deposition model estimates for eastern and northern North America also predict declining fluxes with latitude (*6−8, 44, 45*). The Danish Eulerian Hemispheric model (DEHM) predicts annual Hg deposition with Hg depletion events ranging from >18 $\mu g\ m^{-2}\ y^{-1}$ in eastern North America to 6−12 $\mu g\ m^{-2}\ y^{-1}$ in the Canadian Arctic archipelago (*7*). Miller et al. (*8*) estimated (dry + wet) Hg deposition for rural areas of the northeastern U.S. and adjacent Canadian provinces ranging from about 4 to 30 $\mu g\ m^{-2}\ y^{-1}$ which is good agreement with Hg $\Delta F a_{dj,F}$'s calculated from sediment deposition for this region. Further discussion of model results is given in the Supporting Information.

The agreement between modeled and measured deposition lends additional support to the hypothesis that most Hg deposited from MDEs is revolatilized (*46*). Whereas most of the Hg deposition is predicted to occur over the ocean near sources of Br and BrO (*23*) near shore terrestrial environments could nevertheless receive greater deposition (*46−48*). We examined this question for the 18 Arctic and 2 subarctic lakes in this study that were within 80 km of the ocean. No significant trend was found between Hg FR or with $\Delta F_{adj,F}$ and log distances or square root of the distance, even after omitting the most distant lake (SI Figure S7), suggesting little net effect. To our knowledge this is the most geographically extensive study of Hg fluxes to lake sediments in North America. While a declining trend away from midlatitude emission sources was observed, subarctic and Arctic lakes nevertheless had significant anthropogenic inputs. These observations are consistent with "Hypothesis 2" discussed by Lindberg et al. (*11*) of slow atmospheric oxidation of mercury, slow deposition of reactive mercury emissions, and increasing importance of global sources in remote regions.

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Supporting Information Available

Additional explanatory text, four Tables, and seven Figures. This material is available free of charge via the Internet at http://pubs.acs.org.

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