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Impacts of coal-fired power plants on trace metals and polycyclic aromatic hydrocarbons (PAHs) in lake sediments in central Alberta, Canada

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Abstract

Trace metals and polycyclic aromatic hydrocarbons (PAH) were analyzed in sediment cores from three central Alberta lakes to determine the contributions of local coal-fired power plants to contaminant loadings. In Wabamun Lake, with four power plants built since 1950 within a 35-km radius, sediment concentrations of mercury, copper, lead, arsenic and selenium have increased by 1.2- to 4-fold. Trace metal enrichments were less pronounced in Lac Ste. Anne and Pigeon Lake, situated 20 km north and 70 km south of Wabamun Lake, respectively. Total Hg flux to Wabamun Lake sediments (21–32 μg m^{-2} yr^{-1}) has increased 6-fold since 1950, compared to 2- and 1.5-fold increases in Lac Ste. Anne and Pigeon Lake, respectively, since *circa* 1900. Total PAH flux to surface sediments was 730–1100 μg m^{-2} yr^{-1} in Wabamun Lake, 290–420 μg m^{-2} yr^{-1} in Lac Ste. Anne, and 140–240 μg m^{-2} yr^{-1} in Pigeon Lake. Without adoption of pollution-abatement technology that compensates for increases in generating capacity, continued expansion of coal-burning industry in Alberta will result in increased contaminant deposition, primarily from local sources.

Introduction

Coal-fired power plants are a major source of contaminants to terrestrial and aquatic ecosystems (Nriagu and Pacyna 1988). Among the contaminants released by coal combustion are trace metals such as mercury (Hg), arsenic (As), selenium (Se), lead (Pb), and copper (Cu), and organic compounds such as polycyclic aromatic hydrocarbons (PAHs). Trace metals and PAHs are pollutants of concern in aquatic ecosystems for their persistence, toxicity, and bioavailability (van Brummelen et al. 1998; MacDonald et al. 2000). Of particular concern are bioaccumulative contaminants such as Hg that concentrate in fish tissue and can pose a health risk to human consumers.

Historical increases in the delivery of trace metals and PAHs to remote and urban lakes have been widely attributed to regional or global increases in fossil fuel emissions, rather than local or regional sources (e.g., Galloway et al. 1982; Readman and Mantoura 1987; Fitzgerald et al. 1998; Gevao et al. 1998). Although paleolimnological studies have been undertaken to track past contaminants (e.g., Rose et al., 2004), mercury (e.g., Hynynen et al. 2004) and other metal levels (e.g., Couillard et al. 2004; Boyle et al. 2004), few studies have reconstructed the depositional

histories of trace metals and PAHs in lakes adjacent to coal-fired power plants. Local impacts of coal combustion on sediment quality have been linked to discharge of fly ash and bottom ash slurries, which are enriched in trace metals (Van Voris et al. 1985; Nriagu and Pacyna 1988). PAHs are predominantly emitted from coal-fired power plants in the vapor phase (Revuelta et al. 1999) and thus widely dispersed. However, where particulate emissions are high, local deposition of less volatile compounds can occur (Cheam et al. 2000). Sources and delivery of PAHs to lakes can be approximated qualitatively by comparing PAH indicator ratios to literature values for fossil fuels and biogenic precursors (e.g., Gschwend and Hites 1981; Yunker et al. 2002). Together, coal-related PAHs and trace metals can provide a signature of coal emissions in lake sediments.

Coal is mined and burned to generate the majority of Alberta's electricity. Unlike other provinces, reliance on coal combustion for energy generation is increasing in Alberta. Four power plants in the Wabamun region produce 54% of Alberta's coal-fired power, and emit 65% of the airborne Hg released by industry in the province (CASA 2003). The coal-fired plants in the region include the largest, fourth largest and seventh largest mercury emitters among Canadian power plants (CEC 2005). No previous studies have been performed to quantify impacts of regional coal combustion on contaminant deposition in Canadian prairie lakes. We have examined the concentration profiles of trace metals and PAHs in sediment cores and reconstructed the historical fluxes of Hg and PAHs to three central Alberta lakes, at a range of distances from the concentrated centre of coal-based industry in the Wabamun Lake region. In this way, we are able to determine the effects of emissions from local coal-related industry on the delivery of contaminants to lakes, in the context of possible historical changes in long-range atmospheric deposition.

Description of study sites

Wabamun Lake (53°34′ N:114°35′ W) is a large (surface area = 82 km^2), shallow lake (mean depth = 6.3 m) situated in Alberta's Boreal Mixedwood region. Similar to most lakes in the region, Wabamun Lake has high nutrient concentrations and algal abundance, pH, and specific conductivity (Table 1). Forested portions of the watershed are dominated by trembling aspen (*Populus tremuloides*) and balsam poplar (*Abies balsamifera*), and soils are predominantly grey luvisols. The watershed is highly developed, with over 1300 cottage lots, a railroad, a four-lane highway, agricultural land, two coal strip mines (Whitewood Mine to the north and the Highvale Mine to the south) and two coal-fired power plants (Wabamun and Sundance). Coal strip mines have operated since 1948, although coal was first extracted from underground mines in 1910. The Wabamun and Sundance power plants are situated on the northeast and southeast shorelines, respectively, of Wabamun Lake (Figure 1). The Wabamun plant (capacity = 398 MW) was commissioned in 1956, but the first coal-fired units were not online until 1963. Bottom ash collected from the Wabamun stack is stored in ash lagoons that discharge into Wabamun Lake. The ash lagoons also receive drainage from the Whitewood Mine. The first units of the much larger Sundance plant (2020 MW) were commissioned in 1970. Water from the Sundance cooling ponds is passed through the Wabamun Lake Water Treatment Plant and discharged to Wabamun Lake. The Sundance cooling ponds also received drainage from the Highvale Mine, which up until 1985 had discharged to Wabamun Lake via Beaver Creek. Two additional coal-fired power plants, Keephills (766 MW), commissioned in 1983, and Genesee (820), commissioned in 1989, are located 20 and 35 km, respectively, southeast of Wabamun Lake.

Two nearby lakes (Pigeon Lake and Lac Ste. Anne) were sampled to determine the areal extent of trace metal and PAH deposition in the Wabamun region. Lac Ste. Anne is situated 20 km north, and Pigeon Lake 70 km south, of Wabamun Lake. The Pigeon Lake and Lac Ste. Anne watersheds are similar to Wabamun Lake in soils, vegetation, and water chemistry, but contain neither coal mines nor coal-burning power plants. Both watersheds have extensive agricultural and residential development. Lac Ste. Anne is a smaller, shallower lake, with a larger drainage ratio than either Pigeon Lake or Wabamun Lake, which are similar in lake and catchment morphometry (Table 1).

Materials and methods

In September 2001, using standard gravity coring techniques (Glew et al. 2001), two cores each were collected from Wabamun Lake's East Basin, Lac Ste. Anne (53°37′ N:114°44′ W) and Pigeon Lake (53°01′ N:114°02′ W) (Figure 1) to reconstruct depositional histories of total mercury (THg) and

Table 1. Physical and chemical properties of study lakes and drainage basins.

Lake	Lake area (km^2)	Mean depth (m)	Drainage ratio	Open or agricultural land (% of watershed)	TP ($\mu g\ L^{-1}$)	pH	Secchi depth (m)	COND ($\mu S\ L^{-1}$)
Wabamun Lake	81.8	6.3	3.2	50	30–34	7.9–9.6	1.9–2.3	417
Lac Ste. Anne	54.5	4.8	11.4	30	44–48	8.1–9.0	1.6–2.2	296
Pigeon Lake	96.7	6.2	1.9	46	29–35	8.2–8.6	2.0–2.9	283

Drainage ratio = drainage basin area: lake area; TP, total phosphorus; COND, conductivity; water chemistry data are May–October means over 2 years (1983–1985; Mitchell and Prepas 1990.

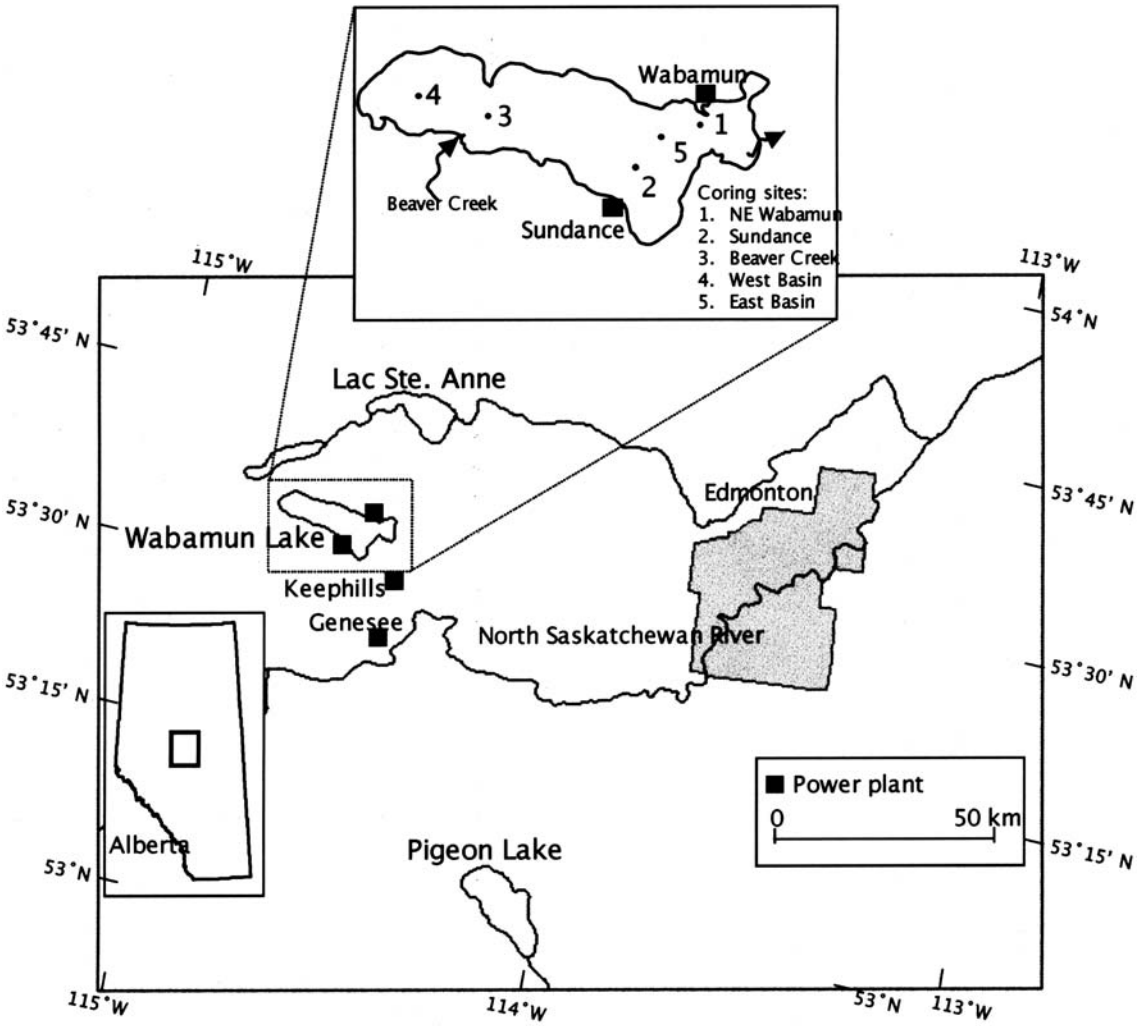

Figure 1. Study area and Wabamun Lake coring sites. The shaded area is Edmonton, a city of about 600,000.

PAHs in these lakes. The cores were relatively short (< 45 cm) and collected from soft sediment, and the gravity corer effectively captured complete post-settlement profiles from each lake. The cores from Pigeon Lake and Lac Ste. Anne also were used for trace metals determinations. In June 2002, additional cores for trace metals analysis were collected from four sites within Wabamun Lake: the northeast portion of the lake adjacent to the Wabamun power plant (NE Wabamun); the southeast part of the lake near the Sundance power plant (Sundance); off the south shore near the Beaver Creek inflow (Beaver Creek); and in the main basin at the west end of the lake (West Basin) (Figure 1). The coring depths in Wabamun Lake were 7 m for the NE Wabamun, Sundance, and East Basin sites, 10 m for the West Basin site, and 11 m for the Beaver Creek site. Cores from Lac Ste. Anne and Pigeon Lake were collected at 9 m.

Each core was divided into 0.5 cm sections over the top 5 cm, and 1 cm sections over the remainder of the core. Sediment age of the cores collected in 2001 was inferred from gamma-counter determinations of ^{210}Pb activity (Appleby 2001) in sediment using the constant rate of supply (CRS) model (Appleby and Oldfield 1978). Activity was measured at the University of Ottawa Department of Biology. Time since deposition was determined by the following equation:

$$t_x = k^{-1} \cdot \ln(A_o \cdot A_x^{-1}) \qquad (1)$$

where k is the decay constant (0.03108 yr^{-1}), A_o is the cumulative inventory of unsupported ^{210}Pb in the core, and A_x is the inventory of ^{210}Pb below depth x. Inventories were calculated with sediment densities based on dry weight and water content of each slice. ^{210}Pb dates were verified with ^{137}Cs activity. Sedimentation rates were calculated for each section of the core as follows:

$$r_x = k \cdot A_x \cdot C_x^{-1} \qquad (2)$$

where C_x is the concentration of $^{210}\mathrm{Pb_u}$ at depth x. Surface flux rates for Hg and PAHs were calculated as the product of the sedimentation rate and contaminant concentration in the upper sections of the East Basin core (Wabamun Lake), the Lac Ste. Anne core, and the Pigeon Lake core. Anthropogenic fluxes were calculated as the difference between 'background' or historical fluxes calculated from the bottoms of the cores and recent fluxes from the top few sections of the cores (Lockhart et al. 1998). Hg and PAH flux rates were corrected for sediment focusing by the focusing factor (FF), calculated as:

$$\mathrm{FF} = f_{^{210}\mathrm{Pbmeasured}} / f_{^{210}\mathrm{Pbexpected}} \qquad (3)$$

where $f_{210\mathrm{Pb\ measured}}$ is the ^{210}Pb flux measured from the sediment cores (dpm m^{-2} yr^{-1}), and $f_{210\mathrm{Pb\ expected}}$ is the expected atmospheric flux of ^{210}Pb based on soil inventories (Lockhart et al. 1998). A range of expected atmospheric fluxes of ^{210}Pb in the Wabamun region was estimated at 0.33–0.48 dpm cm^{-2} yr^{-1}, based on ^{210}Pb soil inventory data from previous studies in northern Alberta and northwestern Canada, respectively (Bourbonniere et al. 1996, Lockhart et al. 1998). We used these two values as a confidence interval to generate focusing-corrected flux rates of Hg and PAH.

Ten to twelve sections per core were analyzed for 45 metals and semi-metals (alkaline earth metals Be, Sr, and Ba; alkali metals Li, Rb and Cs; transition metals Sc, Ti, V, Cr, Co, Ni, Cu, Zn, Y, Zr, Nb, Mo, Ag, Cd, Lu, Hf, Ta, W, and Hg; metals Tl, Pb, and Bi; metalloids As and Sb; rare earth metals La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Th, and U) and 2 non-metals (Se and P). Trace metal samples were freeze-dried at −50 °C and 134 millitor prior to analysis by inductively coupled plasma (ICP) mass spectrometry at the University of Victoria, BC. Detection limits ranged from 0.001 to 0.85 μg g^{-1} except for P (42 μg g^{-1}) and Sr (5 μg g^{-1}). Mean recovery on CANMET standards was 85 ± 4%; replicate samples (n = 5) for each element had a CV of 2 ± 0.3%. Total Hg concentrations in sediment were determined at the Low-level Environmental Mercury Laboratory in the Department of Biological Sciences at the University of Alberta, using cold-vapor atomic fluorescence detection (detection limit = 0.5 ng g^{-1}). Only THg was measured in Wabamun Lake's East Basin core, collected in 2001, whereas all trace metals were measured in the other Wabamun Lake cores from 2002.

Top and bottom sections of cores collected in 2001 from the three lakes were analyzed for 21 PAH compounds: naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, perylene,

benzo(*a*)anthracene (B(*a*)P), benzo(*b*)fluoranthene (B(*b*)F), benzo(*k*)fluoranthene (B(*k*)F), benzo(*e*) pyrene (B(*e*)P), benzo(*a*)pyrene (B(*a*)P), indeno(1,2,3*c,d*)pyrene (IP), dibenzo(*a,h*)anthracene (D(*ah*)A), and benzo(*g,h,i*)perylene (B(*ghi*)P). The depths of top and bottom sections were as follows: Wabamun Lake, top 1 to 2 cm, and bottom 32–33 cm; Lac Ste. Anne, top 1.5–2.5 cm, and bottom 33–34 cm; Pigeon Lake, top 1–3 cm, and bottom 43–44 cm. PAH concentrations (wet weight) were determined by high-performance liquid chromatography (HPLC) with a fluorescence detector at the Freshwater Institute, Department of Fisheries and Oceans, Winnipeg, MB. Detection limits ranged from 0.1 to 2.8 ng g^{-1}. Mean percent recoveries on anthracene *d*10 and terphenyl *d*14 were 100 ± 1.6 and $92 \pm 1.7\%$ ($n = 6$), respectively. Recoveries for SRM2260 averaged $83 \pm 2.6\%$ ($n = 20$). Dry weight PAH and Hg concentrations were derived from wet sample concentrations that were adjusted for water content.

Organic matter content was estimated from loss on ignition (LOI, % dry weight), determined after combustion of sediments at 550 °C for 3 h (Dean 1974). Organic carbon was calculated as:

$$C_{org} = 0.48 \ \ \mathrm{LOI} - 0.73 \qquad (4)$$

where C_{org} is organic carbon concentration (% dry weight; Håkanson and Jansson 1983).

Data analysis

Trace metal concentrations were regressed against sediment depth, to determine significant changes in concentration over time (best-fit curve estimation). Because depth profiles of ^{210}Pb activity indicated sediment mixing in the top ~10 cm of the dated cores, regression analyses were restricted to points below the 10 cm depth. Surface enrichment ratios (SER) for trace metals and PAHs were calculated as the ratio of top:bottom concentrations. Trace metal SERs were calculated from mean concentrations in the top three and bottom three sections of each core; PAH SERs were calculated from concentrations in single top and bottom sections. To account for non-detectable PAHs in total PAH (ΣPAH_{21}) concentrations and fluxes, individual values for "non-detect" (ND) samples were randomly selected from the range of possible concentrations (i.e., 0 to detection level). All statistical analyses were performed on SPSS v. 11.5 and were considered significant at $p < 0.05$.

Results

^{210}Pb profiles and geochronology

When plotted against cumulative sediment mass, the ^{210}Pb curves generally fit an exponential model (Figure 2a–c). However, they were flattened near the top of each core, suggesting bioturbation or physical mixing of surface sediments (Binford et al. 1993). Bottom sections of each core clearly pre-dated the power plants on Wabamun Lake. Peak ^{137}Cs activity (1963) agreed with ^{210}Pb-inferred dates in Lake Ste. Anne, but not in Pigeon and Wabamun lakes where the ^{137}Cs peaks corresponded to ^{210}Pb dates of *circa* 1970 and 1980, respectively (Figure 2). However, ^{137}Cs-inferred dates in the Wabamun and Pigeon cores yield unrealistically low sedimentation rates, thus discrepancies between ^{137}Cs and ^{210}Pb-inferred dates may reflect mobility of ^{137}Cs in the sediment core, rather than an underestimation of dates with ^{210}Pb (Blais et al. 1995). Flux of ^{210}Pb to surface sediments was highest in Lac Ste. Anne, indicating a focusing factor of 1.8 to 2.6 (Table 2).

Recent sedimentation rates (mean ± SE of top three sections) were 90 ± 5 mg cm^{-2} yr^{-1} in Lac Ste. Anne, 40 ± 4 mg cm^{-2} yr^{-1} in Wabamun Lake, and 30 ± 2 mg cm^{-2} yr^{-1} in Pigeon Lake (Figure 3). Sedimentation rates in Pigeon Lake and Lac Ste. Anne briefly peaked *circa* 1930, perhaps reflecting cottage development and conversion of forests to agricultural land of each watershed. Over the last 100 years, sedimentation rates have increased by 4-fold in Wabamun Lake, and 2-fold in each of Pigeon Lake and Lac Ste. Anne.

LOI profiles

LOI in surface sediments of Wabamun Lake at five sites was 28–46% (Figures 4 and 5). Lac St. Anne had a relatively low surface LOI of 20%, compared to 40% in Pigeon Lake LOI (Figure 5). At most sites, LOI decreased to between 20 and 25%

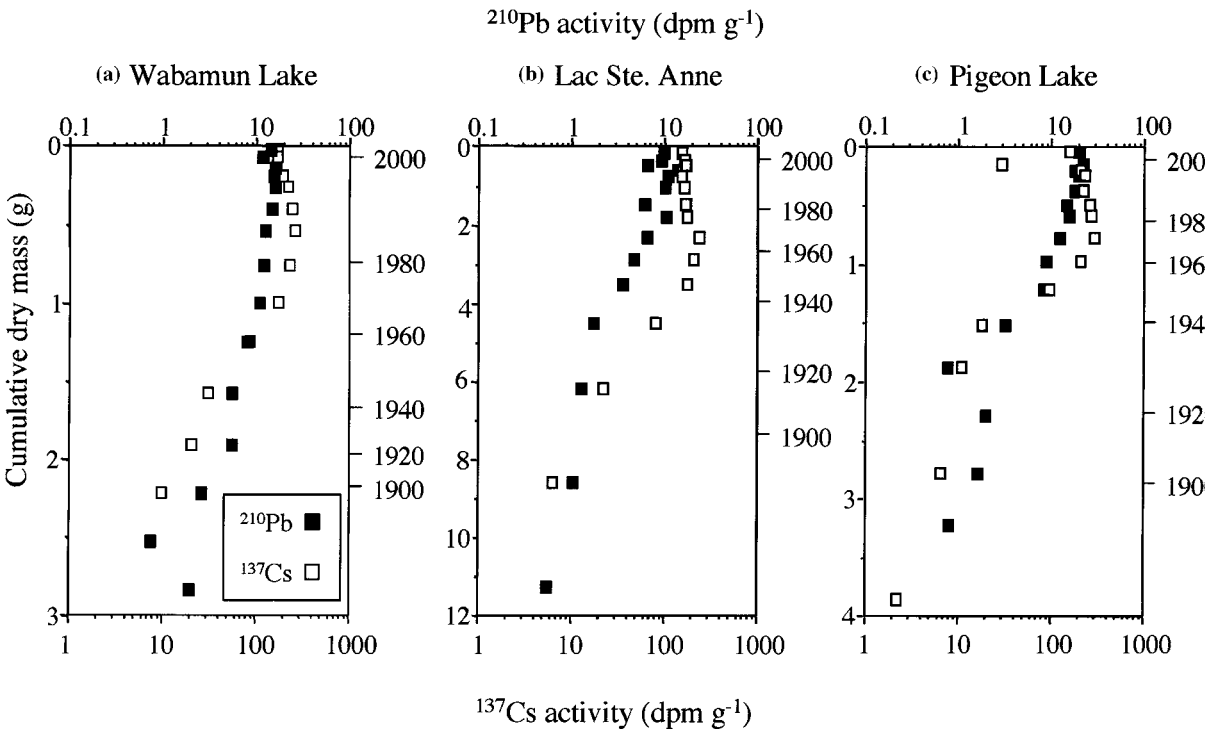

Figure 2. ^{210}Pb and ^{137}Cs activities from (a) Wabamun Lake, (b) Lac Ste. Anne, and (c) Pigeon Lake (dates inferred from ^{210}Pb activity; Wabamun Lake core is from the East Basin site).

Table 2. Sedimentation rates, FFs and ^{210}Pb flux.

Lake	Recent sedimentation rate (mg cm^{-2} yr^{-1})	Surface ^{210}Pb flux (dpm cm^{-2} yr^{-1})	Expected ^{210}Pb flux (dpm cm^{-2} yr^{-1})	FF
Wabamun Lake	40	0.59	0.33–0.48	1.2–1.8
Lac Ste. Anne	90	0.86	0.33–0.48	1.8–2.6
Pigeon Lake	30	0.65	0.33–0.48	1.3–2.0

Recent sedimentation rate calculated from mean of top three sections; expected ^{210}Pb fluxes to the Wabamun region were approximated from Lockhart et al. (1998) (0.48) and Bourbonniere et al. (1996) (0.33); FF = surface ^{210}Pb flux/expected ^{210}Pb flux.

in bottom sediments. In the East and West Basin cores, LOI remained relatively constant throughout the cores, while in Lac Ste. Anne, LOI decreased to 4% in bottom sediments.

Trace metals

Surface enrichments of trace metal concentrations were detected for 10 of 46 elements analyzed in Wabamun Lake, including Hg, Pb, Cu, As and Se (linear or curvilinear regression; $0.50 < r^2 < 0.89$, $p < 0.05$) (Figures 4 and 5). Surface or sub-surface enrichments in Wabamun Lake sediments were also detected for P, Sr, Mo, Sb, and W ($0.54 < r^2 < 0.87$, $p < 0.05$). Trace metal concentrations generally peaked at depths of 10–15 cm, followed by either a gradual decrease in the top 10 cm (e.g., Cu at NE Wabamun; Hg at Beaver Creek; As in West Basin), or a decrease followed by an increase in the top 5 cm (e.g., Hg, Se, As at NE Wabamun site) (Figure 5). SERs of trace metal concentrations ranged from 1.2 to 4.1, although in the NE Wabamun core, mid-core concentrations were up to 10-fold higher than background concentrations (e.g., Cu, Figure 5). The West Basin site in Wabamun Lake, furthest from the two shoreline power-plants, had fewer

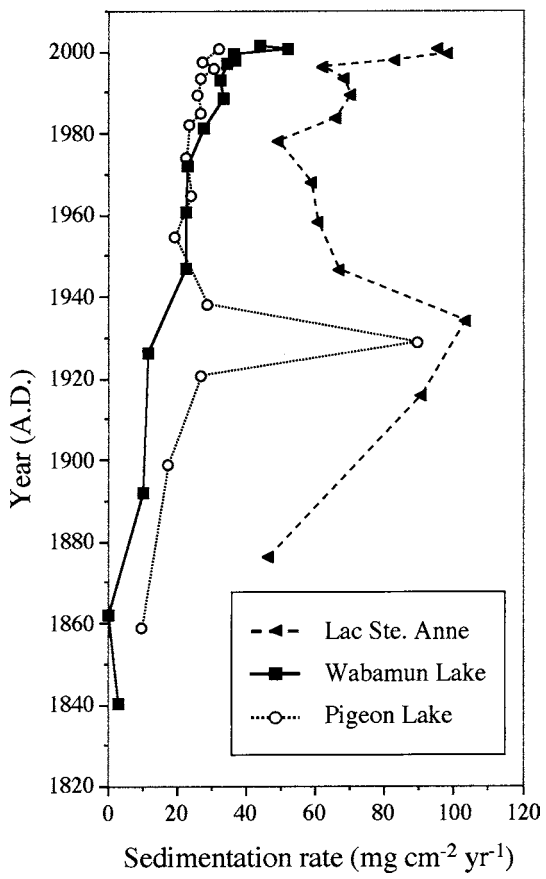

Figure 3. ^{210}Pb inferred sedimentation rates in Wabamun Lake, Lac Ste. Anne, and Pigeon Lake.

detectable trace metal enrichments than NE Wabamun, Sundance, or Beaver Creek, but generally had the highest metal concentrations. Mean concentrations of Hg, Mo, Pb, U, Cu, Zn, Se, Cd, and Sb were higher in the West Basin core than at least 2 of the other three sites in Wabamun Lake (mean of 12 sections per core; ANOVA followed by Tukey's test; $p < 0.05$).

^{210}Pb-inferred dates from the East Basin core indicate that the initial increases in trace metal concentrations at 20 cm coincided with the start-up of the Wabamun power plant in the mid-1950s (Figure 4). The Wabamun cores taken in 2002 (NE Wabamun, Sundance, Beaver Creek and West Basin) were not dated; however, similar to the East Basin, trace metal concentrations in the nearby Sundance and NE Wabamun cores began to increase at the 20 cm depth and thus likely occurred over a similar timeframe.

In Lac Ste. Anne, surface enrichments were detected for 22 of 46 elements analyzed ($0.66 < r^2 < 0.92$, $p < 0.05$), but were generally lower in magnitude than in Wabamun Lake. Similar to Wabamun Lake, surface enrichments of Cu (SER = 2.4), Pb (1.8), and Se (1.8) concentrations were detected in Lac Ste. Anne, and began *circa* 1950 (Figure 6). Other metals that increased in concentration over time in Lac Ste. Anne include Ni (SER = 1.7), Zn (1.6), Cr (1.5), Sb (1.3), and Cs (1.7). Neither Hg nor As increased in concentration between the bottom of the core and the 10 cm depth, nor did they appear to increase over the top 10 cm (Figure 6). Unlike in Wabamun Lake, several of the metals that increased in concentration were rare earth metals (e.g., Dy, Er, Ho, Tb, and Yb), which are not associated with coal emissions.

In Pigeon Lake, 70 km south of Wabamun Lake, only three elements increased in concentration from the bottom of the core: Pb (SER = 1.3), Sr (1.4) and P (2.8) ($0.59 < r^2 < 0.78$, $0.01 < p < 0.04$). The increase in Pb in Pigeon Lake occurred over a similar timeframe to that of Wabamun Lake and Lac Ste. Anne (1950 to 1980; Figure 6). In contrast to Wabamun Lake and Lac Ste. Anne, surface concentrations of Cu and Se in Pigeon Lake decreased in recent sediments.

Mean concentrations of trace metals in the cores were generally higher in Wabamun Lake (composite of four cores) than in Lac Ste. Anne and Pigeon Lake, most notably for Cu (2.7- and 7.1-fold higher, respectively), Mo (4.7- and 12.5-fold), As (3.3- and 1.7-fold), Se (3.4- and 1.7-fold) and Hg (2.7- and 1.9-fold) (ANOVA followed by Tukey's test, $p < 0.001$).

Mercury flux

Focus-corrected flux of Hg to surface sediments in Wabamun Lake was 21–31 μg m^{-2} yr^{-1}, compared to 10–15 μg m^{-2} yr^{-1} in Lac Ste. Anne and 6–9 μg m^{-2} yr^{-1} in Pigeon Lake (Figure 7). Mercury flux in Wabamun Lake increased 7-fold over the last 150 years, compared to a 2-fold increase in Lac Ste. Anne over 180 years, and a 1.5-fold-increase in Pigeon Lake over 100 years. Background Hg fluxes were similar among lakes until 1960, when Hg flux to Wabamun Lake

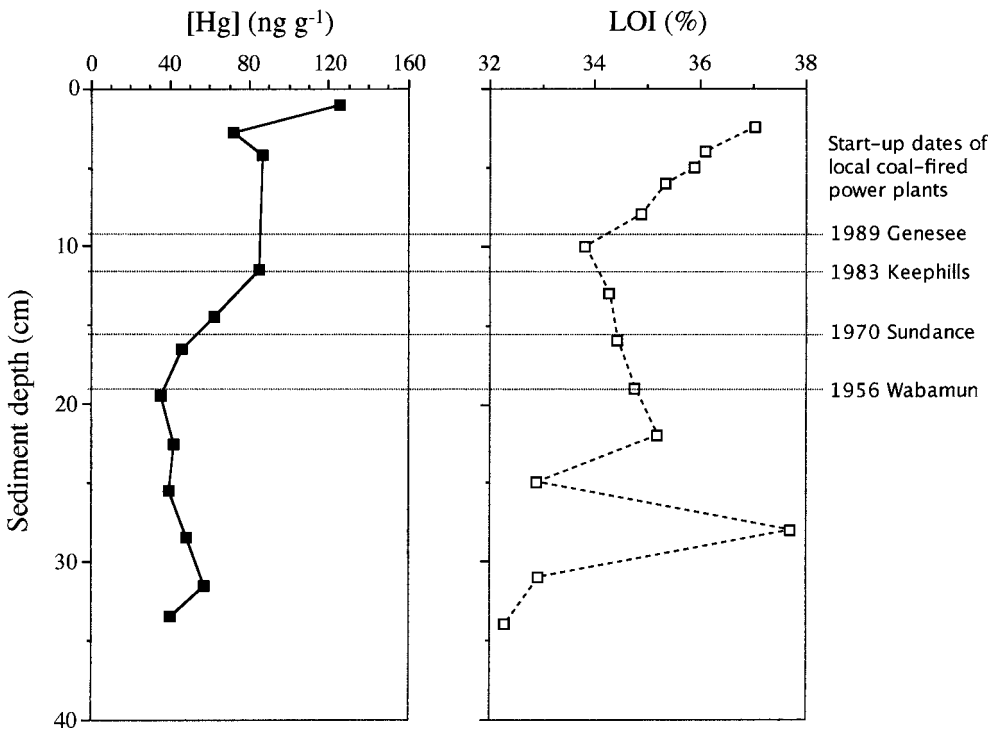

Figure 4. Total mercury concentration and LOI in Wabamun Lake's East Basin core.

began to increase exponentially. From 1956 to 2001, the average annual increase in Hg flux to Wabamun Lake was 3.9%, compared to 1.6% over the 'pre-coal combustion' period of 1840 to 1956. In comparison, Lac Ste. Anne and Pigeon Lake exhibited average annual increases in Hg flux of 1.2% (1818 to 2001) and 0.7% (1915 to 2001), respectively. Anthropogenic Hg flux (recent – historic flux) accounted for 85% of the total flux to Wabamun Lake, and 50% of total flux to Lac Ste. Anne and Pigeon Lake (Tables 3 and 4). Thus Hg flux has increased to the greatest extent in Wabamun Lake, and decreases with increasing distance from the four power plants surrounding it.

PAHs

Twenty of 21 PAHs were detected in the surface sediments of Wabamun Lake for a total concentration of 2756 ± 4 ng g^{-1}, compared to 771 ± 10 ng g^{-1} in Lac Ste. Anne (19 PAHs detected) and 895 ± 166 ng g^{-1} in Pigeon Lake (10 PAHs detected). After correction for organic carbon, total PAH concentrations were still elevated in Wabamun Lake (16,306 ± 24 ng g organic C^{-1}) relative to Lac Ste. Anne (8211 ± 106 ng g organic C^{-1}) and Pigeon Lake (4766 ± 883 ng g organic C^{-1}). Total PAH concentrations in deep sediments were also elevated in Wabamun Lake (448 ± 61 ng g^{-1}; 11 PAHs detected) relative to Lac Ste. Anne (62 ± 20 ng g^{-1}; 8 PAHs detected) and Pigeon Lake (213 ± 41 ng g^{-1}; 9 PAHs detected).

Total PAH flux to surface sediments was 730–1100 μg m^{-2} yr^{-2} in Wabamun Lake, 280–420 μg m^{-2} yr^{-1} in Lac Ste. Anne, and 100–240 μg m^{-2} yr^{-1} in Pigeon Lake (Figure 8). Fluxes of individual PAH compounds have increased in Wabamun Lake by 35-fold since 1850, compared to 54- and 20-fold increases in Lac Ste. Anne and Pigeon Lake, respectively, since *circa* 1800. The corresponding average annual increases (accounting for differences in timeframe among the cores) were 2.3% in Wabamun Lake, 2.0% in Lac Ste. Anne, and 1.5% in Pigeon Lake. While these data suggest a higher rate of annual increase in PAH flux to Wabamun Lake than in the other two lakes, it should be noted that many of the PAHs

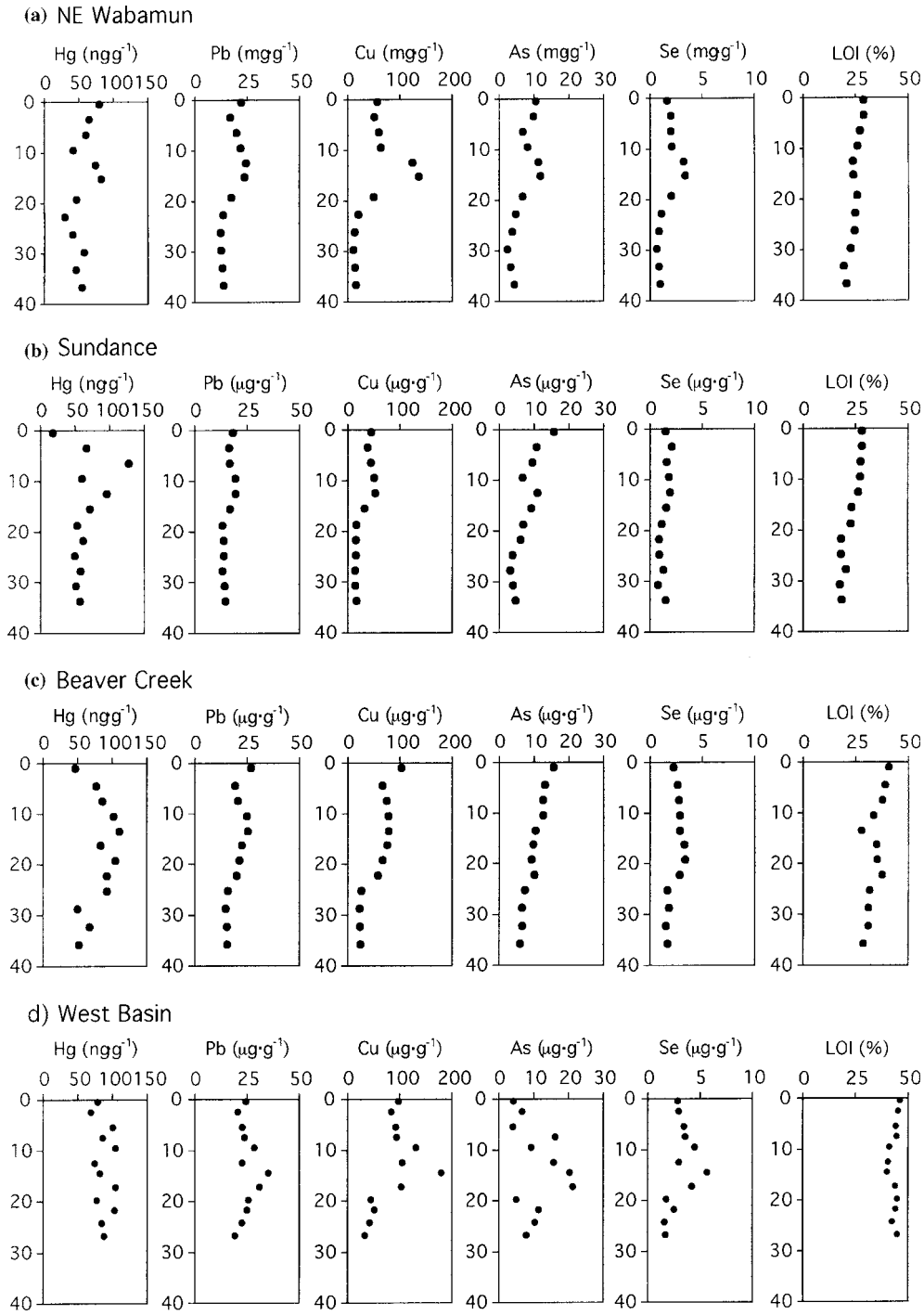

Figure 5. Sediment profiles of selected trace elements from Wabamun Lake sites: (a) NE Wabamun, (b) Sundance, (c) Beaver Creek, and (d) West Basin.

that exhibited surface enrichments were undetectable in bottom sediments. Therefore SERs and annual increase rates for the three lakes represent minimum estimates, based on the substitution of detection limits for actual concentrations in bottom sediments.

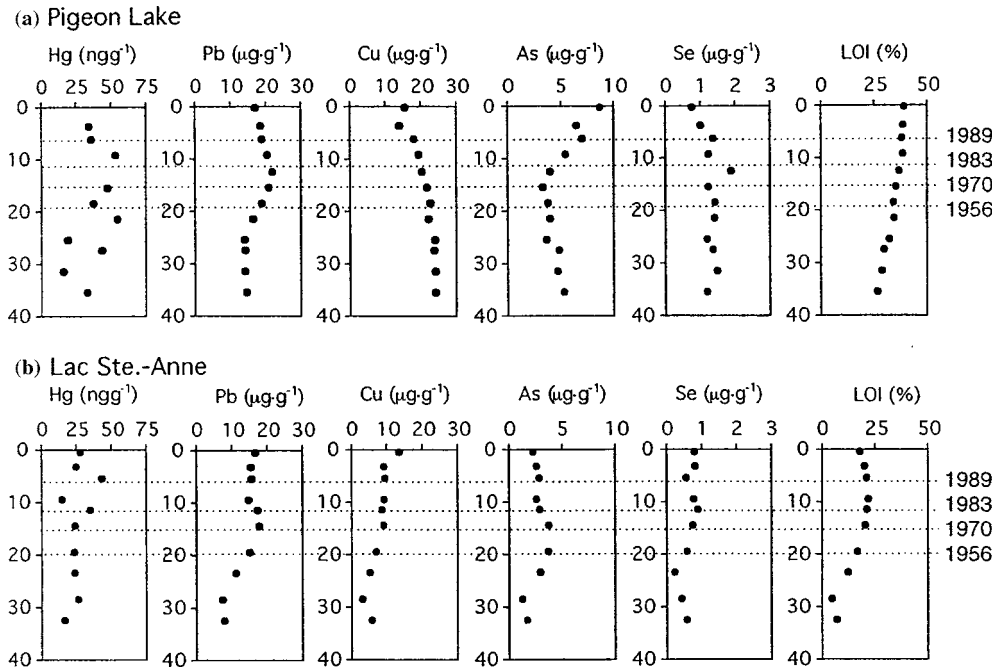

Figure 6. Sediment profiles of selected trace metals from (a) Pigeon Lake and (b) Lac Ste. Anne (dates indicate start-up years for nearby coal-fire power plants).

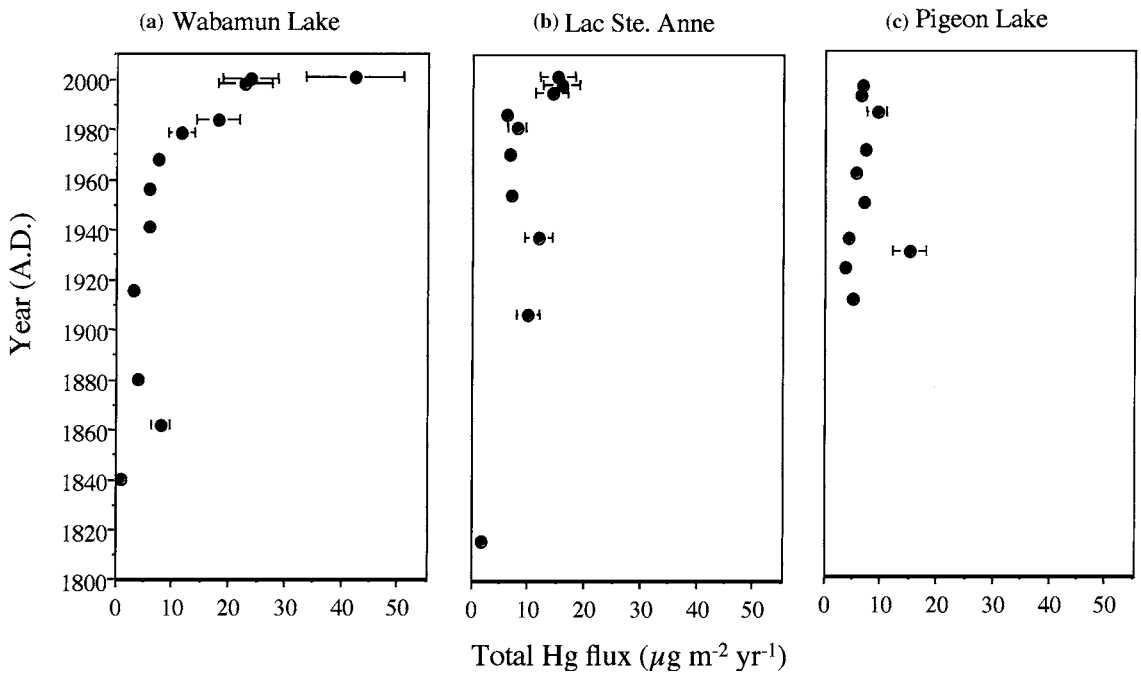

Figure 7. Mercury flux rates in (a) Wabamun Lake, (b) Lac Ste. Anne and (c) Pigeon Lake (bars represent the confidence interval generated by sediment FFs).

Table 3. Mercury and PAH fluxes.

Lake	Recent flux			Historical flux			Anthropogenic flux (μg m^{-2} yr^{-1})
	(μg m^{-2} yr^{-1})	Timeframe	*n*	(μg m^{-2} yr^{-1})	Timeframe	*n*	
Hg							
Wabamun Lake	21–32	1984–2001	4	3–5	1840–1880	3	18–27
Lac Ste. Anne	10–15	1986–2001	4	5–7	1820–1910	2	5–8
Pigeon Lake	6–9	1986–1997	3	3–5	1910–1920	2	3–4
ΣPAH_{21}							
Wabamun Lake	730–1100	2001	1	28–42	1850	1	700–1060
Lac Ste. Anne	290–420	1999–2000	1	8–12	1800	1	410–630
Pigeon Lake	160–240	1998–2000	1	6–13	1800		150–230

Anthropogenic flux = recent flux – historical flux; *n* refers to number of core sections used to calculate fluxes.

Shifts in the relative abundance, or percent composition, of PAHs between bottom and top sections were qualitatively similar between Wabamun Lake and Lac Ste. Anne cores. In Wabamun Lake, the relative abundance of the PAHs associated with fossil fuel combustion [i.e., IP, B(*ghi*)P, B(*a*)P, B(*e*)P, B(*b*)F, B(*k*)F, chrysene, B(*a*)A, fluoranthene, and pyrene; see Prahl and Carpenter 1983] increased from 36% in deep sediments to 59% at the surface. In contrast, the relative abundance of perylene, a biogenic compound, decreased from 26% in deep sediments to 15% in surface sediments. In Lac Ste. Anne, the relative abundance of combustion PAHs increased from 42 to 59%, and perylene decreased from 20 to 9%, between bottom and top sections of the core. Thus anthropogenic, fossil-fuel-derived PAHs have increased in the surface sediments of Wabamun Lake and Lac Ste. Anne, relative to the dominant natural PAH compound.

In Pigeon Lake, only three of the 10 principal combustion PAHs were detected in surface sediments (chrysene, pyrene, and fluoranthene), and their relative abundance decreased from 47% in deep sediments to 32% in surface sediments. Similar to Wabamun Lake and Lac Ste. Anne, perylene decreased in relative abundance from 15 to 5% between deep and surface sediments. In contrast to the other lakes, the Pigeon Lake core exhibited an increase in the relative abundance of low molecular-weight compounds (naphthalene, 1-methylnaphthalene, and 2-methylnaphthalene), from 29% at the base of the core to 42% at the surface. The relative abundance of these compounds decreased slightly between deep and surface sediments in Wabamun Lake and Lac Ste. Anne. Recent increases in PAH loading to Pigeon Lake are dominated by volatile, 2-ring compounds, whereas Wabamun Lake and Lac Ste. Anne have received greater inputs of high molecular-weight, 4- and 5-ring compounds typical of coal and gasoline combustion.

Discussion

Trace metals

Surface enrichment of trace metal concentrations and flux in Wabamun Lake are indicative of long-term increases in deposition from local coal-related industry, rather than global sources. Increased concentrations of trace metals associated with coal combustion, such as Hg, Cu, Pb, As, Sb, Sr, Mo, and Se, coincided with the establishment of coal-fired power plants and other industries in the Wabamun Lake watershed. Increased loadings of Cu and As have resulted in surface sediment concentrations that exceed "lowest effects levels" (LEL) for toxicity to benthic organisms (MacDonald et al. 2000).

The Wabamun Lake results are consistent with recent studies that demonstrate enrichments of fly ash and trace metals in lake and river sediments near coal-fired power plants (e.g., Ravi Chander et al. 1994; Cheam et al. 2000). Despite fluctuations in trace metal concentrations in the upper sediments of Wabamun Lake, our Hg flux calculations indicate that loading rates have increased over the past 50 years. Although add-on pollution controls (i.e., electrostatic precipitators

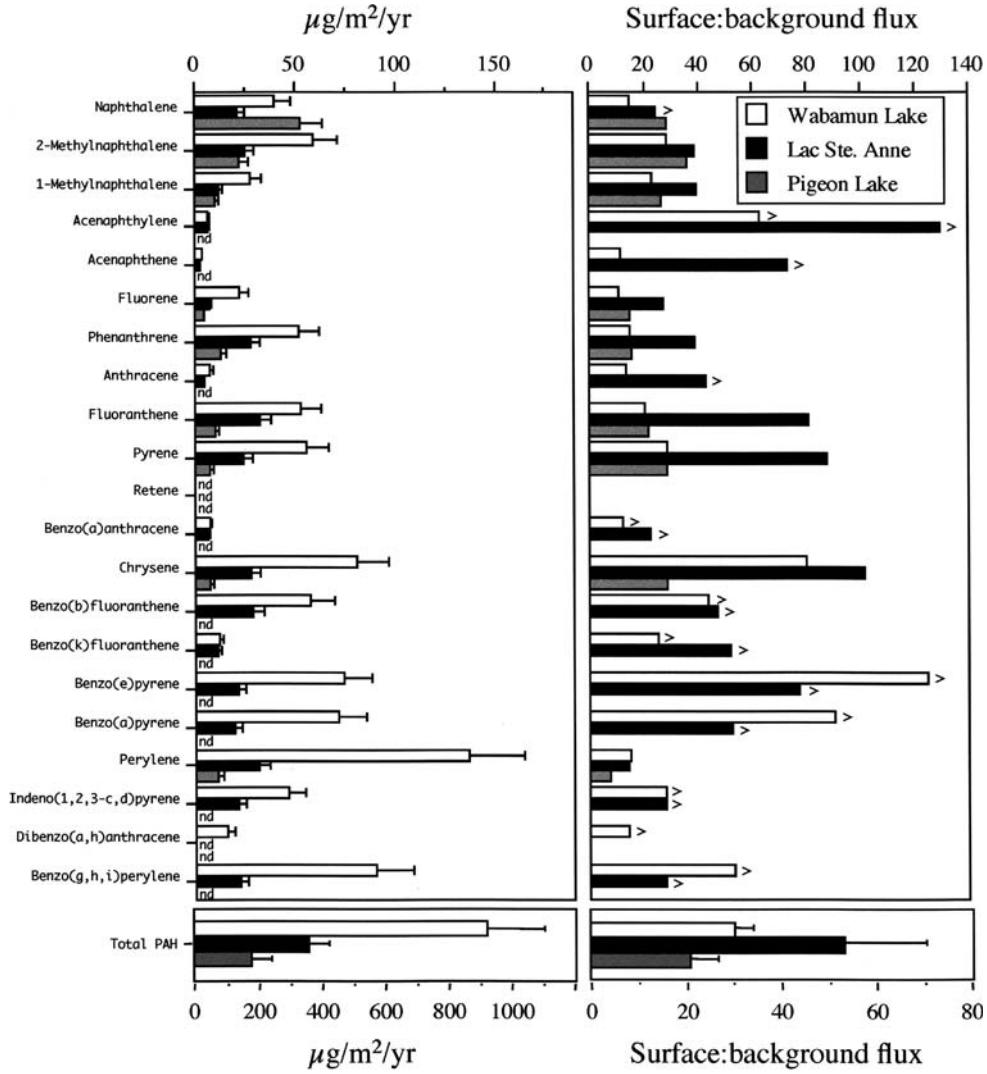

Figure 8. (a) Surface PAH flux and (b) SER ('>' indicates that the ratio is greater than or equal to the indicated value, due to non-detectable PAH concentration in bottom sediments; error bars indicate the range for total fluxes and ratios, reflecting the range of possible concentrations for undetectable compounds (i.e., 0 to detection limit) and estimates of FFs).

and fabric filters) have been implemented at Wabamun-area power plants, recent particulate emissions amount to over 6000 t per year (1999 data; SENES Consultants Ltd. 2002). The Sundance plant alone discharged 35 t of Pb, 12 t of Cu, 8 t of As, and 0.4 t of Hg in 2000 (NPRI 2001), representing the dominant source of trace metals in the region and the largest point-source emitter of Hg in Canada (CEC 2005). While declines in trace metal concentrations after improvements to fly-ash controls have been documented elsewhere (Kirby et al. 2001), large increases in coal-burning capacity in the Wabamun Lake area appear to have overwhelmed any emission reductions that might have followed adoption of pollution-control technologies.

Relatively low trace metal concentrations and fluxes in Lac Ste. Anne and Pigeon Lake suggest that coal-related industry has a localized impact on trace metal loadings to lakes in the Wabamun region. Van Voris et al. (1985) found that particulate emissions from a coal-fired power plant in Battle River, Alberta were deposited within a 50 km radius, with major deposition of ash

particles occurring 2–5 km from the power plant. In Lac Ste. Anne, situated 20 km north of Wabamun Lake, enrichments of Cu, Se, and Pb suggest that the lake receives some fallout from the coal-fired power plants to the south. Surface enrichments of rare earth metals in Lac Ste. Anne, however, were more indicative of watershed erosion than coal combustion. Pigeon Lake, up to 70 km south of the power plants, is likely outside the range of particulate fallout, with the possible exception of the Genesee station (Figure 1). Prevailing winds also are in a NW–SE direction, and Pigeon Lake likely receives low atmospheric deposition from the power plants to the north. Sanei et al. (2001) found surface enrichments of trace metals such as As, Cu, Zn, and Cr in Pigeon Lake sediments after normalizing for the clastic mineral detritus fraction, but suggested that industrial inputs were minimal when compared with natural processes of erosion.

Trace metal profiles in the Wabamun Lake cores were further distinguished from those of Lac Ste. Anne and Pigeon Lake by mid-core enrichments, particularly at the NE Wabamun site adjacent to the Wabamun power plant. The NE Wabamun core exhibited mid-core peaks in Hg, Pb, Cu, As, and Se concentrations between ^{210}Pb-inferred dates of 1950 and 1980. Flux of these metals to the NE Wabamun site may have been enhanced by surface inputs from an industrial gravel washing operation that discharged to the lake over a 20-year span from the 1950s to the mid-1970s. A recent survey of the surface sediment quality in the gravel washing canal found high concentrations of Al, As, Fe, Sn, V, and U, relative to shallow sediments in an adjacent bay (Golder Associates Ltd. 2002). The same study reported high concentrations of As, Cr, Cu, Mo, and Se at littoral sites in Wabamun Lake near the ash lagoon discharge (NE Wabamun).

The West Basin exhibited the fewest detectable trace metal enrichments of the Wabamun cores, but had peak concentrations of Cu, Pb, As, Sb that were among the highest of the Wabamun sites, possibly reflecting high organic matter content (see Forstner 1987). Trace metal profiles from the West Basin were likely affected by lake-wide surficial sediment mixing and redistribution processes, given the sampling site was at the deepest point of this wide, shallow bay. The potential for sediment focusing at the site may have contributed to high sedimentation rates, necessitating a deeper core from the West Basin to capture depositional histories over the same timeframe as the other sites.

We acknowledge that there are difficulties in extrapolating trace metal loadings from concentration data. Other potential sources for the fluctuations in trace metal concentrations in upper sediments could include dilution of metal concentrations by recent increases in sedimentation rate (e.g., Engstrom et al. 1994), redirection of mine drainage inputs from Beaver Creek to the Sundance cooling pond, and disruption of surface sediments from substantial macrophyte harvesting operations.

It has been suggested elsewhere that surface enrichments of trace metals, including Hg, may be the artifact of diagenesis and post-depositional redistribution (Rasmussen et al. 1998). However, productive lakes are less likely to be affected by diagenetic processes due to low decomposition rates (Matty and Long 1995), indicated in Wabamun Lake by gradual declines in LOI. In a recent review on Hg geochemistry, Fitzgerald and Lamborg (2003) concluded that sediment Hg is largely unaffected by post-depositional redistribution processes. Furthermore, other metals that are not redox-sensitive species, including Mo, As, and Se (Forstner 1987), exhibited surface enrichments in Wabamun Lake similar to that of Hg. Trace metal profiles in Wabamun Lake clearly deviated from LOI profiles, indicating that surface enrichments were the result of increases in metals deposition from local coal-based industry.

Mercury flux

Mercury flux to Wabamun Lake clearly reflects a local source, rather than increased atmospheric deposition of background, globally sourced Hg. Mercury flux to remote and rural lakes in North America has typically increased by 2- to 4-fold over the last 150 years (Fitzgerald et al. 1998), whereas Hg flux to Wabamun Lake has increased 7-fold over the same period, with the major increase occurring after the commencement of coal-fired power generation in the Wabamun area. The increase in Hg flux to Wabamun Lake is comparable to 5- to 7-fold increases observed in European lakes situated near industrialized regions

(Iverfeldt 1991). The relatively modest increases in Hg loading observed in Pigeon Lake and Lac Ste. Anne are more similar to Hg trends in remote lakes of northwestern Canada, where sediment Hg fluxes increased by 1.1- to 1.6-fold in the last 100–150 years (Lockhart et al. 1998). Paleoreconstruction of Hg fluxes in Lake Athabasca in northern Alberta also suggested no long-term increases (Bourbonniere et al. 1996), further implicating local sources of Hg to lakes in the Wabamun area, rather than increases in regional or global deposition.

Mercury flux rates determined for our study lakes were generally within the range of values reported in the literature. Background (pre-1900) Hg flux in our study lakes (3–5 μg m^{-2} yr^{-1}) was similar to values reported in lakes from remote northern Minnesota and Wisconsin (Engstrom et al. 1994), northwestern Ontario (Johnson 1987) and northwestern Canada (Lockhart et al. 1998). However, recent anthropogenic Hg flux to Wabamun Lake was above the range of values reported by these studies. The increased Hg flux in Wabamun Lake is most likely a combined result of atmospheric deposition of particulate Hg released from local coal-fired power plants, and surface inputs related to watershed development and erosion, including extensive coal mining.

PAHs

Coal and gasoline combustion appear to be the principal sources of PAHs to Wabamun Lake and Lac Ste. Anne, and to a lesser extent, Pigeon Lake. Flux and relative abundance of parent compounds typical of fossil fuel combustion, such as acenaphthylene, fluoranthene, pyrene, chrysene, B(*e*)P, B(*a*)P, and B(*ghi*)P (Howsam and Jones 1998; Simcik et al. 1999; Prahl and Carpenter 1983), increased by 1–2 orders of magnitude in the study lakes over 150–200 years. Combustion PAHs are not subject to major degradation in anaerobic sediments (Readman and Mantoura 1987), therefore sediment core profiles should accurately reflect historical changes in PAH inputs. In Wabamun Lake, the largest increases in flux occurred among the high molecular-weight, 4-, 5- and 6-ring compounds, B(*e*)P, B(*a*)P, chrysene, and B(*ghi*)P. The heavier, 5- and 6-ring compounds are generally associated with the particulate fraction of fossil fuel emissions, especially gasoline combustion (Howsam and Jones 1998). Their high relative abundance in Wabamun Lake may reflect the proximity of the lake to major emission sources other than coal-fired power plants, such as local highway, boat, and industrial traffic. Revuelta et al. (1999) noted an absence of 5- and 6-ring PAHs in the emissions of Spanish coal-fired power plants, and a predominance of lower molecular-weight, 3- and 4-ring compounds in the vapor phase, notably pyrene, phenanthrene, chrysene, and fluoranthene. While the flux of these compounds also increased in Wabamun Lake, the increases were less pronounced than those of the 5- and 6-ring PAHs, suggesting a lesser impact of coal emissions relative to other fuels. Alternatively, poorly controlled particulate emissions from the Wabamun area power plants may result in higher emissions of heavy PAHs than detected in the study by Revuelta et al. (1999).

In Lac Ste. Anne, the largest individual increases in PAH flux occurred among the lighter, 3- and 4-ring compounds (i.e., acenaphthylene, fluoranthene, pyrene, and chrysene), identified by Revuelta et al. (1999) and others as markers of industrial coal combustion (see Howsam and Jones 1998). Accumulation of these compounds in Lac Ste. Anne may reflect their higher vapor pressures and wider dispersal relative to the heavier 5- and 6-ring compounds that were abundant in Wabamun Lake. This assertion is supported by our findings that the largest increases in fluxes in Pigeon Lake, the most distant lake from coal-combustion sources in the Wabamun region, were of the volatile, 2-ring compounds, naphthalene, 1-methylnaphthalene, and 2-methylnapthalene. Fluxes of chrysene and pyrene increased to a lesser extent, and 5- and 6-ring compounds were undetectable in Pigeon Lake's surface sediments. While the abundance of alkylated naphthalene derivates in Pigeon Lake are indicative of a petrogenic source (i.e., unburned fossil fuel), increases in the delivery of PAHs to the study lakes were generally dependent on the distance of the lakes from the power plants and the volatilities of individual compounds.

Total and individual PAH concentrations and fluxes in the study lakes were comparable to values in remote lakes, but several orders of magnitude lower than values reported from urban or industrialized regions of North America and Europe

(e.g., Gschwend and Hites 1981; Wickstrom and Tolonen 1987; Gevao et al. 1998). This is attributable to the rural setting and the lack of major industrial centres or urban regions upwind of the study lakes. However, increases in PAH flux (i.e., the difference between deep and surface sediments), particularly for Wabamun Lake and Lac Ste. Anne, were higher than values reported from remote and rural sites and comparable to some urban sites (e.g., Gschwend and Hites 1981; Readman and Mantoura 1987; Gevao et al. 1998). Sedimentary records from rural and urban sites typically show a peak in total PAH flux between 1930 and 1960 followed by a decline, reflecting reductions in coal as a home heating fuel and improvements in combustion technology (Gevao et al. 1998; Howsam and Jones 1998). In contrast, coal-related industry in the Wabamun region has expanded at the rate of one power plant (1000 MW) per decade over the past 40 years, negating possible emissions reductions via improved combustion technologies, and resulting in relatively large increases in SERs for PAH flux. Despite these large increases, however, PAH concentrations remain well below LELs for sediment biota (MacDonald et al. 2000).

As with trace metals, PAH concentrations and fluxes in Wabamun Lake, Lac Ste. Anne, and Pigeon Lake indicate a predominantly local impact from coal-based industry and other sources in their watersheds, and suggest that more distant sources of PAHs such as the city of Edmonton or long-range transport have not greatly impacted lakes in this region. This lack of increase in long-range anthropogenic impacts is also reflected in Lake Athabasca, where PAH flux has been dominated by natural sources since 1900 and has corresponded to regional trends in forest fire activity (Bourbonniere et al. 1996). Surface concentrations of individual PAHs also were 5- to 25-fold higher in Wabamun Lake than in Lake Athabasca. However, this may be partly explained by the low total organic matter of Lake Athabasca sediments (1–2%) relative to Wabamun Lake (14%). The lack of surficial increase in PAH concentrations in Lake Athabasca suggests that local anthropogenic sources are likely the primary contributors to the trends observed in the Wabamun region.

Qualitative PAH indicator ratios suggest that the majority of PAHs in the study lakes were derived from combustion, but offered seemingly contradictory results as to the source fuels (Table 4). For example, B(*e*)P:[B(*e*)P + B(*a*)P] ratios for both Wabamun Lake (0.51) and Lac Ste. Anne (0.52) are closest to gasoline emissions (0.53) (Grimmer et al. 1985), coal-fired residential furnaces (0.48) (Grimmer and Hildebrandt 1975), and hard coal combustion (0.43) (Ratajczak et al. 1984). However, several other ratios support a strong coal combustion signal in Lac Ste. Anne. For example, fluoranthene:(fluoranthene + pyrene) ratios in Lac Ste. Anne (0.57) are closest to those of hard coal emissions (0.58) (Ratajczak et al. 1984), whereas a lower ratio in Wabamun Lake (0.49) is perhaps more indicative of automobile emissions (0.43) (Grimmer and Hildebrandt 1975). Similarly, IP:[IP + B(*ghi*)P] ratios for Wabamun Lake (0.34) are intermediate between coal soot (0.56) (Cretney et al. 1985) and gasoline emissions (0.18), whereas the value for Lac Ste. Anne (0.49) is more indicative of coal

Table 4. PAH indicator ratios from study lakes and potential sources.

PAH ratio	Site			Source			
	Wabamun Lake	Lac Ste. Anne	Pigeon Lake	Coal emissions	Gasoline emissions	Coal soot	Crude oil
Fl/(Fl + Py)	0.49	0.57	0.58	0.58[a]	0.43[b]	–	0.22[e]
IP/(IP + B(*ghi*))	0.34	0.49	–	0.48–0.57[e]	0.18[c]	0.56[c]	0.09[e]
B(*e*)P/(B(*e*)P + B(*a*)P)	0.51	0.52	–	0.43[a], 0.48[f]	0.53[b]	–	–
Phen/fluorene	2.4	4.2	2.9	4[d]	–	–	–
Phen/Fl	0.98	0.86	1.3	3.5[d]	–	–	–
Fl/fluorene	2.4	4.8	2.3	1.5[d]	–	–	–

Fl, fluoranthene; Py, pyrene; IP, indeno(1,2,3-*c*,*d*)pyrene; B(*e*)P, benzo(e)pyrene; B(*a*)P, benzo(*a*)pyrene, Phen, phenanthrene; [a]Ratajczak et al. (1984), [b]Grimmer and Hildebrandt (1975), [c]Cretney et al. (1985), [d]Revuelta et al. (1999), [e]Yunker et al. 2002, [f]Grimmer et al. (1985).

combustion. Lastly, the phenanthrene:fluorene ratio in Lac Ste. Anne (4.2) is more similar to the ratio from power plant emissions (4) by Revuelta et al. (1999) than either Wabamun Lake (2.4) or Pigeon Lake (2.9).

These sorts of qualitative ratios to indicate PAH emission sources are based on large sample numbers. Equally, they are typically used with large datasets and to identify general trends, with substantial range in site-specific values. Our use of only two samples per lake prevents us from drawing strong conclusions of source-dominance based solely on these qualitative PAH ratios. Given that all three study lakes have substantial recreational development and are very popular recreational destinations, increased PAH deposition in central Alberta lakes is likely due to emissions from coal processing and coal-fired power plants, as well as other local fossil fuel-based sources.

Conclusions

Surface enrichments of sedimentary trace metals and PAHs in Wabamun Lake are indicative of fossil fuel combustion, and implicate adjacent coal-fired power plants as dominant sources. The close coincidence of increases in Hg deposition within Wabamun Lake-with construction and operation of four coal-burning power plants and two coal surface-mines in the area, and diminishing deposition with distance, suggests a causal relationship between coal-related industry in the area and increased Hg deposition. Continued reliance on coal-generated power and expansion of coal-related industry in Alberta, without adoption of emissions controls that compensate for massive capacity increases, will result in continued increases in trace metal and PAH deposition on the regional landscape. While there currently is evidence of limited local sediment quality problems associated with trace metals, this also can be expected to increase with anticipated future expansion of coal-combustion in Alberta. As has been seen elsewhere in the world, these trends of increasing contaminant deposition are likely reversible, provided appropriate pollution-control technologies are adopted that reduce absolute amounts of point-source emissions.

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