Fluxes of As, Cu, Hg, Pb in lake sediments in the Coppermine River basin, Canada

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Abstract Many watersheds in northern Canada are experiencing increasing pressures from resource extraction, development and the long-range transport of atmospheric pollutants. This study examines sediment accumulation and the spatial and temporal distribution of trace metals in bottom sediment of down gradient lakes in the headwaters of the Coppermine River basin, Canada. Sediment cores were collected from Lac de Gras, Desteffany Lake, Point Lake and Daring Lake using a plastic lined K–B single-gravity corer. Each core was dated using ^{210}Pb and concentrations of trace metals (As, Cu, Hg, Pb) were determined in core sections. Sedimentation rates ranged from 101 $\mathrm{g\,m^{-2}\,yr^{-1}}$ at Desteffany Lake to 156 $\mathrm{g\,m^{-2}\,yr^{-1}}$ at Daring Lake and are comparable to other northern lakes. Concentrations of As and Cu were significantly higher at Lac de Gras. Metal loading data and enrichment ratios show that concentrations of Pb and Hg are elevated compared to historic background levels. Metal enrichment is from anthropogenic activities and atmospheric inputs. Lake sediment represents a good indicator of state for the Coppermine basin and documents historic trends of metal deposition. However, the indicator has low sensitivity to change and coarse temporal resolution due to low sedimentation rates in northern environments.

Keywords Coppermine River; environmental indicator; lake sediment; sediment cores; trace metals

Introduction

Scientific knowledge regarding the state of northern aquatic ecosystems is incomplete (Landers *et al.* 1995) and ecological uncertainty in these environments is high (Mulvihill and Jacobs 1991). Consequently, gaps in scientific knowledge have serious implications for planning and management of northern ecosystems. The Coppermine River basin is a transboundary watershed located in the western portion of the Northwest Territories and Kitikmeot Region of Nunavut, Canada. The basin is typical of many remote northern ecosystems experiencing resource extraction and development pressures causing environmental change (MacDonald *et al.* 1999). Water resource managers are concerned about ecosystem health of the basin and the sensitive northern aquatic ecosystems are significant to aboriginal culture and health (Cizek *et al.* 1995). Consequently, there is a need to develop suitable indicators to assist in environmental planning and management of aquatic ecosystems in the Canadian north, including the Coppermine River basin.

Lake sediment is one potential environmental indicator for planning and managing northern ecosystems. The quality of bottom sediment influences the health of aquatic ecosystems (McIntosh 1991) and integrates terrestrial and atmospheric inputs to lakes, thus providing an indicator of the degree of pollution in aquatic ecosystems (Kumar *et al.* 1998). Bottom sediment contains a historical record of environmental information (Baudo *et al.*

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1990) and analyses of metals in sediment cores have been used to determine natural, background conditions and accumulation of metals from anthropogenic sources (Engstrom *et al.* 1994) including the long-range transport of atmospheric pollutants (Lockhart *et al.* 1998). While a few studies have investigated surface characteristics of lake sediment in the Coppermine River basin (MacDonald *et al.* 1999; Stephens 1999; Puznicki 1997), little is known about historical changes in sediment accumulation and changes to metal loading in the basin. This paper examines the distribution of trace metals (As, Cu, Hg, Pb) in sediment cores collected from a series of down-gradient lakes in the Coppermine River basin. Sediment cores are dated to estimate sedimentation rates and infer historical changes to metal loading.

Methods

Study area and sample collection

The Coppermine River basin is a trans-boundary watershed located in the western portion of the Northwest Territories and Kitkimeot Region of Nunavut, Canada (Figure 1). The basin drains an area of 50 800 km^2 and the Coppermine River flows 845 km north from the headwater region to its confluence with the Arctic Ocean at Kugluktuk (Wedel *et al.* 1988). The Coppermine River basin is undergoing significant land use change due to resource extraction and concerns have been raised about the cumulative effects of mining and other human activities on water quality and the health of northern aquatic ecosystems (MacDonald 1999). Diamond mining activities are ongoing in and near Lac de Gras, at the headwaters of the river.

Sediment cores were collected from four study lakes in the headwaters region of the Coppermine River basin; Lac de Gras, Desteffany Lake, Point Lake and Daring Lake (Table 1). Lac de Gras, Desteffany Lake and Point Lake form a down-gradient chain of long, narrow lakes. Diamond mining is the predominant land use in the headwaters region of the Coppermine River basin. Daring Lake is the smallest of the study lakes and it receives tributary inflow from Yamba Lake. Daring Lake is designated as an Environmental Monitoring and Assessment Network (EMAN) site and there is currently, no mining activity in this basin. Subsequently, Daring Lake was used in this study as a reference site in which to compare environmental data from the other three study lakes.

Sediment cores were collected at sites with the maximum lake depth using a stainless steel Kajak–Brinkhurst single-gravity corer with plastic liners (Mudroch and Azcue 1995). These deep water sites incorporate inputs from all portions of the lake and can provide an uninterrupted sedimentary sequence to provide information about the entire lake (Charles *et al.* 1994). Cores were sectioned at 1 cm intervals in the top 10 cm and at 2 cm intervals below 10 cm. The lake surface area, core location, water depth and core length for each lake is shown in Table 1.

Laboratory methods and calculation of enrichment ratios

Sediment cores were dated using ^{210}Pb at MyCore Scientific, Deep River, Ontario. The ^{210}Pb profiles from the study lakes show the cores have an uninterrupted sedimentary sequence which is required to investigate temporal trends in sediment characteristics. Decreasing ^{210}Pb levels with depth show little disturbance over time and that relatively undisturbed sediment profiles were obtained from each lake (Peramaki 2001). Sedimentation rates were estimated according to the constant rate of supply model (Engstrom *et al.* 1991).

In this study, four trace metals (As, Cu, Hg, Pb) are evaluated because they have both environmental significance (i.e. may pose a threat to aquatic biota) and associated sediment quality guidelines (CCME 1999; OME 1993). Sediment from each core section was freeze dried. Concentrations of Cu and Pb were determined at the Taiga Environmental Laboratory,

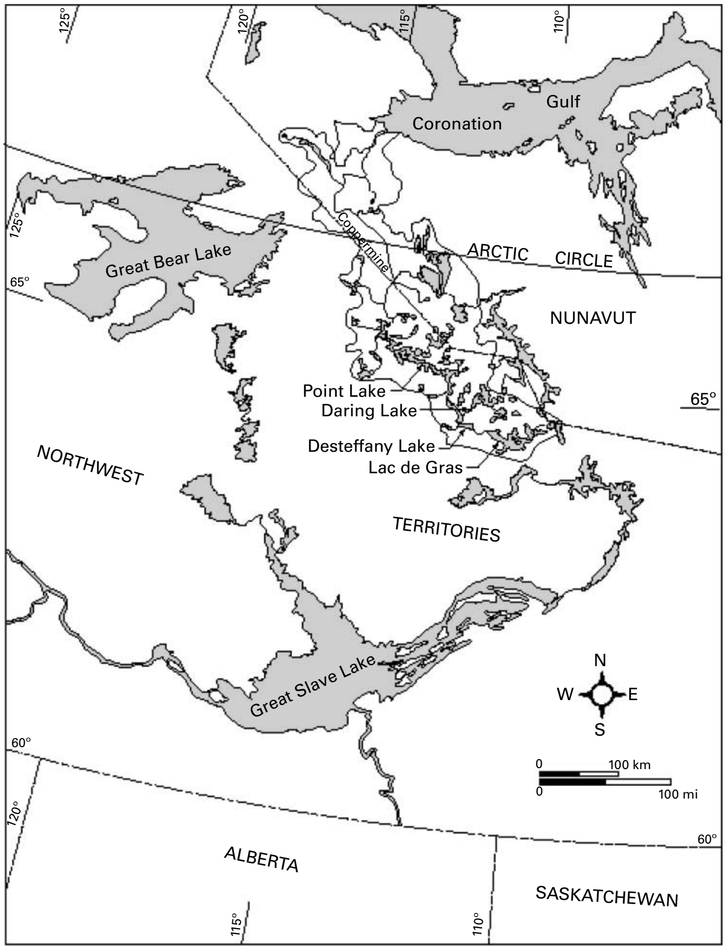

Figure 1 Study area

Yellowknife, NWT using ICP-MS according to USEPA Method 200.8. Arsenic was analyzed by AAS using the hydride technique (APHA Method 3114) and Hg by AAS using the cold vapour technique (GSC 1973). Accuracy of the analysis was verified by running Canadian reference standards and comparing results with stated reference values for trace elements (Lynch 1990). Reproducibility of the analyses was good and the relative standard deviation was <5% for Cu, Hg and Pb and <9% for As.

Metal enrichment factors were determined by comparing recent and background metal concentrations (Lockhart *et al.* 1995). Enrichment ratios (ER) are defined as the ratio of surface (recent) to background metal fluxes. Surface flux is calculated by multiplying sedimentation rates with metal concentrations in the surface sediment section (0–1 cm depth).

Table 1 Location and characteristics of sediment cores

Lake	Lake area (km^2)	Latitude	Longitude	Water depth (m)	Core length (cm)
Lac de Gras	577	64° 32′ N	110° 58′ W	30	28
Desteffany Lake	40	64° 36′ N	111° 36′ W	55	28
Point Lake	594	65° 17′ N	113° 05′ W	60	28
Daring Lake	15	64° 50′ N	111° 38′ W	27	30

The background metal flux is estimated by multiplying metal concentrations from 10–12 cm depth with median sedimentation rates for the core. Metal enrichment ratios <2 are interpreted as inputs primarily from natural, geologic sources within the watersheds while enrichment ratios >2 indicate inputs primarily from anthropogenic sources (Lockhart *et al.* 1995). Metal profiles were also compared to Al profiles. Metal profiles with patterns visually similar to the Al profile are considered to be largely influenced by natural phenomena, while metals that increase at a rate faster than that of Al may be anthropogenic pollutants (Gubala *et al.* 1995).

Results and discussion

Sedimentation rates

Mean sedimentation rates in the four study lakes are generally low; 101 g m^{-2} yr^{-1} at Point Lake, 130 g m^{-2} yr^{-1} at Lac de Gras, 141 g m^{-2} yr^{-1} at Desteffany Lake and 156 g m^{-2} yr^{-1} at Daring Lake. The higher sedimentation rates in Daring Lake are likely due to suspended solids input from tributary inflow draining Yamba Lake. While these rates may be overestimated because sediment was sampled in deep-water sites (Charles *et al.* 1994), they are comparable to other northern lakes, including Great Slave Lake (54–112 g m^{-2} yr^{-1}) (Mudroch *et al.* 1989) and high Arctic lakes (121–278 g m^{-2} yr^{-1}) (Muir *et al.* 1995). In contrast, sedimentation rates in the four study lakes are lower than in Lake Ontario, a temperate lake (325–635 g m^{-2} yr^{-1}) (Kemp and Thomas 1976).

Metal distribution

In As, Cu, Hg and Pb profiles, metal concentrations increase from the bottom to the top of the core (Figures 2–5). In Slipper Lake in the Coppermine River basin (Stephens 1999), concentrations of As and Cu are generally lower at Desteffany, Point and Daring Lakes but levels of Hg are comparable. Metal concentrations in Lac de Gras are often higher than concentrations previously reported in surficial sediment by MacDonald *et al.* (1999).

National sediment quality guidelines have been developed to interpret possible effects of sediment-associated metals in aquatic ecosystems (CCME 1999). The Interim Sediment Aquatic Guideline (ISGQ) indicates a level that likely has no effect on the majority of sediment dwelling organisms. The Probable Effects Level (PEL) indicates a level that is

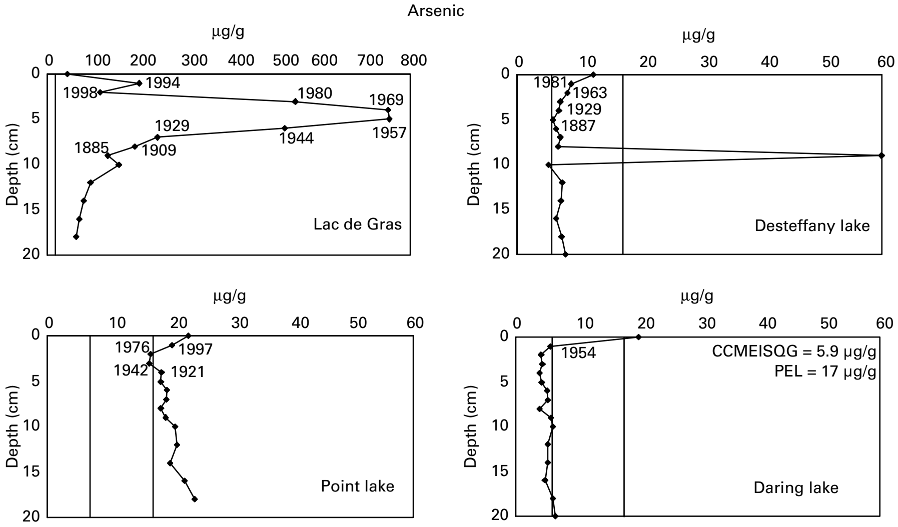

Figure 2 Arsenic profiles

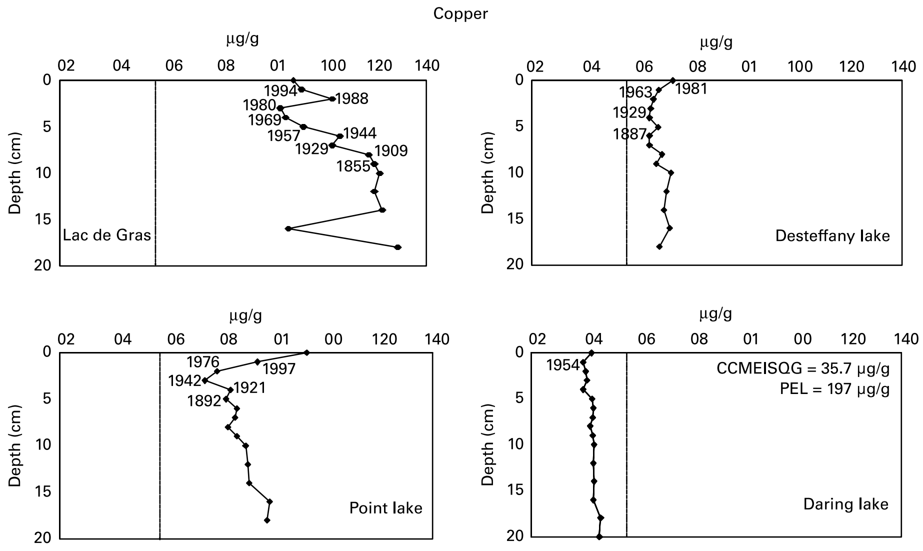

Figure 3 Copper profiles

likely to affect aquatic biota adversely. Metal (As, Cu, Hg, Pb) concentrations in dated sediment cores are compared to the ISGQ and PEL of the CCME aquatic sediment guidelines in Figures 2–5, respectively. Lead and Hg concentrations of sediment in the four study lakes are below the ISQG guidelines for the protection of aquatic life and therefore pose little risk to biota (Figures 4 and 5). Levels of Cu are above the ISQG in Lac de Gras, Desteffany and Point Lake (Figure 3). The PEL for As is exceeded in surface sediment collected from Lac de Gras, Point Lake and Daring Lake (Figure 2).

Metal enrichment and loading

Previous studies have reported metal enrichment in bottom sediment of northern Canadian lakes. Lead and Hg were enriched in southeastern Hudson Bay by a factor of 3.2 and 3.5,

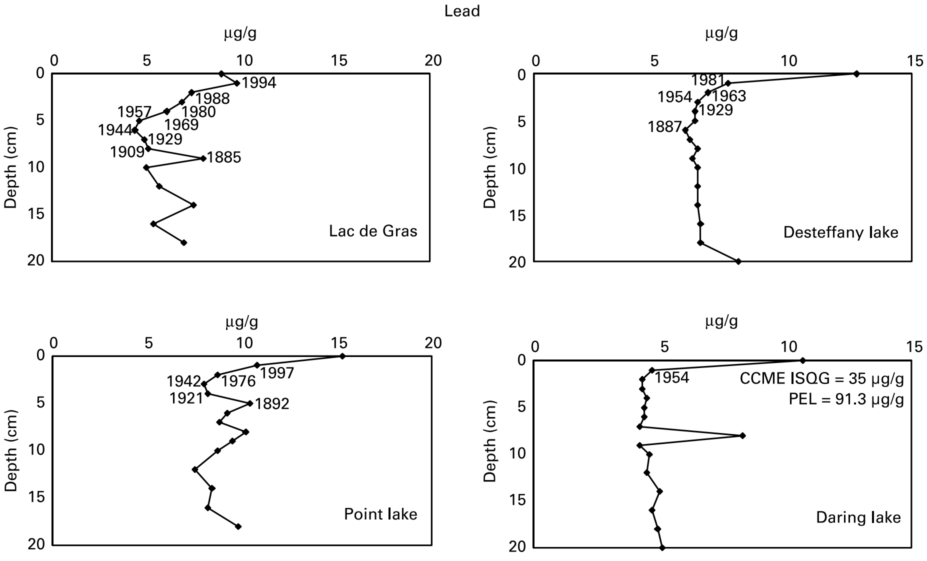

Figure 4 Lead profiles

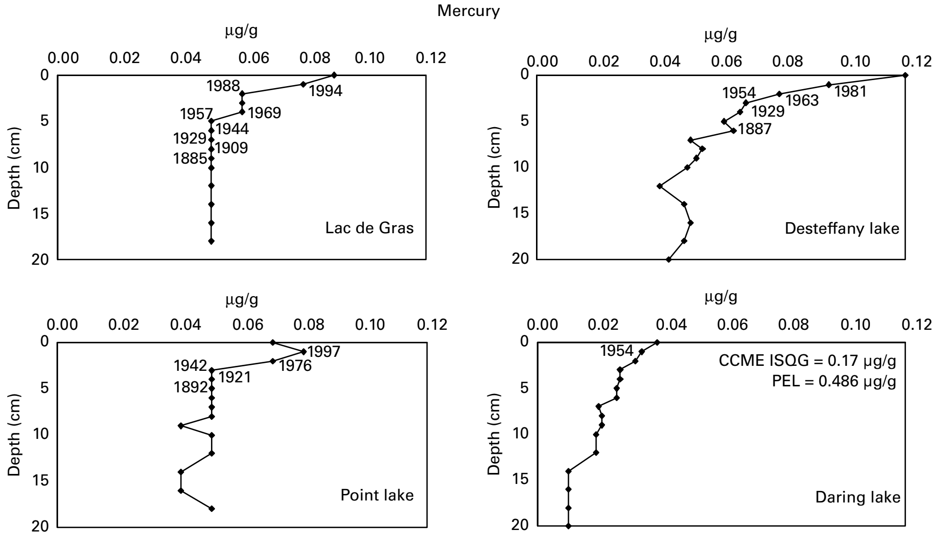

Figure 5 Mercury profiles

respectively (Hermanson 1991). In northern Quebec lakes, Lucotte *et al.* (1995) reported enrichment in Pb by factors of 3.0 to 147 and Hg by factors of 2.3 to 11. Metal enrichment has been reported in Norwegian and Russian Arctic lakes where concentrations of Pb are enriched by a factor of 1 to 6, while Hg is enriched by a factor of 1.6 to 2.8 (Rognerud *et al.* 1998). All three studies attributed the enrichment of metals in lake sediment to anthropogenic processes, specifically long-range transport of atmospheric pollutants (LRTAP).

Metal loading and enrichment were variable for lakes in the Coppermine River basin. According to criteria developed by Lockhart *et al.* (1995), metal loading at Point Lake is dominated by natural weathering of surficial materials (i.e. natural, geologic sources). All metal enrichment ratios at Point Lake are <2 and metal loading from anthropogenic sources is considered to be very low. While Pb and Hg are slightly enriched in surface sediment at Point Lake, natural weathering is most likely the primary source of these metals. Loadings of metals have increased, but have not exceeded geologic inputs (Lockhart *et al.* 1995). This finding is supported by comparisons of Pb and Hg with Al profiles, which suggest input of materials from the natural landscape (Peramaki 2001).

Other lakes in the Coppermine basin show evidence of recent metal inputs dominated by anthropogenic sources. At Desteffany and Daring Lakes, Hg enrichment ratios are greater than or equal to 2. Changes in metal flux suggest an increase in Hg loading from anthropogenic sources (Lockhart *et al.* 1995). Surface fluxes range from 6.2 to $12.6\,\mu g\,m^{-2}\,yr^{-1}$ and historic fluxes range from 3.0 to $6.2\,\mu g\,m^{-2}\,yr^{-1}$. Mercury enrichment at Lac de Gras may also be attributed to anthropogenic sources (ER = 1.9). Comparisons of Hg and Al profiles at Lac de Gras, Desteffany and Daring Lakes also suggest that Hg enrichment is related to anthropogenic sources.

At Daring Lake, the Pb enrichment ratio is >2, which indicates an increase in Pb loading likely from anthropogenic sources (Lockhart *et al.* 1995). Surface flux of Pb is $1712\,\mu g\,m^{-2}\,yr^{-1}$ and the historic flux is $718\,\mu g\,m^{-2}\,yr^{-1}$. Lead enrichment at Lac de Gras may also be attributed to anthropogenic sources (ER = 1.9). Comparisons of Pb and Al profiles at these two lakes indicate anthropogenic sources have influenced Pb loading.

The source of anthropogenic metal enrichment in the study lakes is most likely LRTAP. Increased metal inputs from anthropogenic sources began a few decades ago. For example, Pb at Daring Lake increased after 1954, while Hg at Desteffany and Daring Lakes increased after 1887 and 1954, respectively. No point sources existed near these lakes at these times and the only possible anthropogenic source is LRTAP. The only other possible explanation for high ER in Daring Lake is the possible impact of sediment-associated metals entering Daring Lake from a tributary draining the much larger Yamba Lake. Yamba Lake basin is the focus of considerable mining exploration and the possibility of metal loading to Daring Lake from the Yamba Lake outflow should be addressed in future studies. Other studies report similar metal enrichment in surface sediment in northern aquatic ecosystems and attributed it to LRTAP (Hermanson 1991; Lockhart *et al.* 1995; Lucotte *et al.* 1995; Rognerud *et al.* 1998).

The implications of metal enrichment for biotic health have not been fully established. However, if some portion of the increased metal levels in sediment enters the aquatic food chain, then increased metal uptake by fish might be expected (Lockhart *et al.* 1995). Previous research has found sediment-associated metal concentrations correlated to metal concentrations in aquatic organisms (Rognerud and Fjeld 1993; Johnson 1987). The pathways of metal transfer from sediment to biota include direct contact with sediment particles and ingestion of sediment particles and sediment-pore water. In the study lakes, enriched metals, particularly As, Pb and Hg, may be detrimental to biota.

While numeric sediment quality guidelines may offer a proxy for potential toxicity, guidelines have not been developed specifically for northern environments and current guidelines may not be applicable in some cases (Puznicki 1997). Consequently, a comprehensive assessment of bottom sediments using the Sediment Quality Triad (Chapman 1990) should be conducted to measure impacts of metals in lake sediments on biota. The Sediment Quality Triad includes sediment chemical analysis, examination of *in situ* benthic community composition and measurement of sediment toxicity. In addition, bioaccumulation measurements should be conducted using lake sediment to determine whether metals are bioavailable, if there is a measured response and if the metals are causing the response (Borgman *et al.* 2001).

Metal enrichment of lake sediment does not decrease with distance from diamond mining activities. However, the low sedimentation rates at the study lakes mean there is substantial uncertainty assessing recent impacts, such as those associated with diamond mining. Nevertheless, Lac de Gras, the focus of mining activities, does not show greater metal enrichment over time compared with other lakes. The source of anthropogenic metal loading to the study lakes is atmospheric transport, rather than point source. Metal enrichment and metal loading in the study lakes is lower than at lakes in northeastern US (Engstrom *et al.* 1994), northern Quebec (Lucotte *et al.* 1995) and southeastern Hudson Bay (Hermanson 1991). Results of the present study are similar to the spatial patterns of contaminant distribution in northern freshwater lakes (Blais and Kalff 1993). There is a tendency for bottom sediment to be less contaminated at higher latitudes and there is a general decrease in metal contamination from east to west in the north, which is likely related to predominant wind patterns (Schindler *et al.* 1995).

Conclusions

The spatial and temporal distribution of trace metals in bottom sediment of down-gradient lakes in the headwaters of the Coppermine River basin was examined to evaluate the role of bottom sediment as an environmental indicator for environmental planning and management of northern aquatic ecosystems. Sedimentation rates in the Coppermine basin are comparable to other lakes in northern Canada. Concentrations of As and Cu were significantly higher at Lac de Gras. Metal loading data and enrichment ratios show that concentrations of Pb and Hg are elevated compared to historic background levels due to atmospheric inputs (rather than point sources). Lake sediment represents a good indicator of state for the Coppermine basin and documents historic trends of metal deposition. The indicator has low sensitivity to change and coarse temporal resolution due to low sedimentation rates in northern environments.

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