

Rationale, research approach, and study sites for investigating vertical metal distributions in lacustrine sediments

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Abstract: Metals such as iron are generally more concentrated near the sediment–water interface than in deeper lake sediments. These observations have been widely reported. This enrichment has been attributed to increased recent anthropogenic deposition and/or to vertical diagenetic remobilization. The Geological Survey of Canada’s Metals in the Environment (GSC MITE) Initiative has established a second phase of lake-sediment research to explore the relative effects of chronological metal loading and diagenetic metal remobilization in lake-sediment profiles.

Two acidic kettle lakes in the Canadian boreal forest, Lake Perron and Lac de la Pépinière, were selected within the zone of influence of the Horne smelter in Rouyn-Noranda (Quebec). Major selection criteria were indications that the sites have been exposed to smelter emissions, i.e. diatom evidence of increased lake-water acidity over time, geographic orientation in the prevailing wind directions with respect to the smelter, and no obvious metal loading from local anthropogenic sources other than the Horne smelter. Since changes in oxidation state and solubility can drive metal mobility, lakes were chosen with a fully oxygenated water column in summer and a high probability of hypolimnetic anoxia in winter.

The rationale behind this study is to assess whether or not lake sediments from the study sites can be used as reliable archives of historical metal loading. The research approach and field sites chosen form essential contextual data from which emerging multidisciplinary analytical results of diatom studies, pollen counts, solid- and aqueous-phase geochemistry, isotopic determinations, bacterial enumerations, and quantitative mineralogy can be interpreted and integrated.

Résumé : Les métaux comme le fer sont généralement davantage concentrés près du point de contact entre les sédiments et l’eau qu’au sein de sédiments lacustres plus profonds, ce qui a été largement observé. Cet enrichissement a été attribué au dépôt récent et plus important de matériaux anthropiques ou à une remobilisation diagénétique verticale ou à ces deux phénomènes. Dans le cadre du programme des Métaux dans l’environnement (MEDE) de la Commission géologique du Canada, on a entrepris une seconde phase de travaux de recherche sur les sédiments lacustres qui visait à étudier les effets relatifs de la remobilisation diagénétique des métaux et des charges en métaux au fil du temps dans les profils de sédiments lacustres.

On a choisi d’étudier deux lacs de kettle acides de la forêt boréale canadienne, soit le lac Perron et le lac de la Pépinière, qui se trouvent dans la zone d’émissions de la fonderie Horne, à Rouyn-Noranda (Québec). Leur sélection était en grande partie fondée sur des indices montrant qu’ils ont été exposés aux émissions de la fonderie, soit des diatomées témoignant d’une augmentation de l’acidité de l’eau lacustre au fil du temps, une orientation dans la direction des vents prédominants par rapport à la fonderie et l’absence de charges en métaux manifestement issues de sources anthropiques autres que la fonderie. Puisque des changements d’oxydation et de solubilité peuvent entraîner une mobilité des métaux, on a choisi des lacs dont la colonne d’eau est totalement oxygénée pendant l’été et fort probablement sujette à l’anoxie hypolimnique durant l’hiver.

Le fondement de cette étude repose sur la détermination que les sédiments lacustres des lacs à l’étude peuvent ou non renseigner de manière fiable sur l’histoire des charges en métaux. L’approche de recherche et les lacs choisis ont fourni les données contextuelles nécessaires à l’interprétation et à l’intégration de résultats tirés d’études sur les diatomées, de numérations polliniques, d’analyses géochimiques des phases solide et aqueuse, de datations isotopiques, de numérations bactériennes et d’analyses minéralogiques quantitatives et obtenus grâce à de nouvelles techniques d’analyse multidisciplinaires.

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INTRODUCTION AND RATIONALE

Reconnaissance surveys of 99 lakes in Phase I of the GSC MITE lake-sediment studies have demonstrated that metal concentrations are elevated in surface lake sediments proximal to the point source (i.e. the Horne smelter in Rouyn-Noranda, Quebec) and decrease with distance in the path of the prevailing wind directions, presumably as a function of anthropogenic loading and distance of transport (Kliza and Telmer, 2001; Bonham-Carter et al., 2005). Additionally, concentrations of metals are higher in surface sediments (0–2 cm) than in deeper sediments (18–20 cm; likely pre-industrial). Similar patterns of metal enrichment in surface sediments have been widely reported in the literature (e.g. Nriagu et al., 1982; Evans et al., 1983; Nriagu and Rao, 1987; Borgmann et al., 2001).

The rationale for studying vertical metal distributions in lacustrine sediments is to improve interpretations of geochemical profiles in undisturbed lake bottoms. The vertical metal distributions observed in sediment cores may not necessarily represent chronological metal deposition (e.g. Farmer, 1991; Friske, 1995; Boyle, 2001). Results of this study will help identify the causes for elevated metal concentrations near the sediment–water interface, i.e. increases in loading from recent anthropogenic activity and/or natural processes of postdepositional redistribution of metals within the sediment column. Diagenetic metal mobility, which overprints the historical archive of chronological metal loading in lake sediments, can raise doubts in paleolimnological reconstructions, although in some environments and for some metals, the historical record appears to be well preserved (e.g. Rae and Parker, 1996; Callender, 2000; Alfaro-De la Torre and Tessier, 2002). Further investigation is required to determine for which metals and under what conditions — environmental, physical, or geochemical — the chronological record of metal loading is preserved and those conditions under which it is confounded by metal mobility. The objectives of this study are 1) to combine traditional and advanced analytical techniques in order to identify the processes that distribute or redistribute metals in lake sediments; 2) to characterize the pre-industrial lake-sediment record as well as lake response to mining and smelting operations; and 3) to evaluate the effects of 1) and 2) in two lakes that have different geochemical settings but are located at a similar distance away and in the prevailing wind directions from a major anthropogenic point source of metals.

A greater understanding of the processes that lead to metal enrichment in surface lake sediments will aid both government and industry in selecting appropriate risk assessment and risk management strategies to minimize deleterious ecosystem effects. Furthermore, this understanding will assist the evaluation of geochemical results from surface lake sediments for exploration purposes (i.e. potential false anomalies). Consequences of high metal concentrations near the sediment–water interface include potential risks to lake-water quality, increased toxic metal availability for biotic uptake, and implications for the use of lake bottoms as long-term storage repositories for acid-generating waste rock.

This paper summarizes the rationale for investigating vertical metal distributions in lake sediments at two pilot sites that are likely receptors of acidic deposition from a major metal point source in the Abitibi-Témiscamingue region. The multidisciplinary research approach that is outlined here offers a comprehensive integrated strategy to assess geochemical profiles of lacustrine sediments as historical records of metal loading. Although lake-sediment research is commonplace, the criteria and process for study lake selection is not always provided; in this paper, a discussion of study site selection is explicitly presented. Finally, geological, physical, and geochemical characterizations of the two study sites are presented to provide a contextual framework for current investigations.

RESEARCH APPROACH

Metals in lake sediments can be liberated to the aqueous phase by diagenetic processes such as reductive dissolution. Conversely, metals in sediment pore water or in the overlying water column can be sequestered either transiently or as a permanent sink into the solid phase (e.g. by adsorption or co-precipitation; Hamilton-Taylor and Davison, 1995). In this study, a wide range of analytical tools are being used to examine both the solid and aqueous phases in order 1) to establish exposure to anthropogenic loading at the study sites (e.g. increased sulphur loading from historical SO₂-rich smelter emissions and resultant increased lake-water acidification); 2) to investigate potential mechanisms of metal transport within the sediment column (e.g. diffusion and bacterially mediated reduction-oxidation reactions); and 3) to compare two lake settings seasonally in order to study the interlake and temporal variability of vertical metal transport in lacustrine sediments.

The scientific approach involves a range of multidisciplinary techniques and measurements launched by the GSC MITE Initiative with partners in both federal government departments and academia (Fig. 1). The approach includes assessing the history of industrial and environmental conditions with evidence from records of smelter feed and emissions, sediment dating techniques, bio-indicators, and stable isotopes. The vertical distribution of metals and their potential mobility and/or transformation are being investigated with evidence from the geochemistry of the solid and aqueous metal pools, the presence and activity of bacterial populations, and the geochemical and mineralogical composition of potential carrier phases for metals (both organic and inorganic).

Documented changes in the operations of the Horne mine and smelter in Rouyn-Noranda can be used to interpret geochemical vertical distributions in dated lake sediments as a function of changes in industrial outputs (see Bonham-Carter, 2005, Table B1). Sediment dating (e.g. ²¹⁰Pb, pollen marker horizons) can verify that the retrieved sediment cores penetrated deeper than the initial record of industrialization in the 1920s (A.S. Dixit, S. Alpay, S.S. Dixit, and J.P. Smol, unpub. rept., 2004). Sediments that predate industrialization

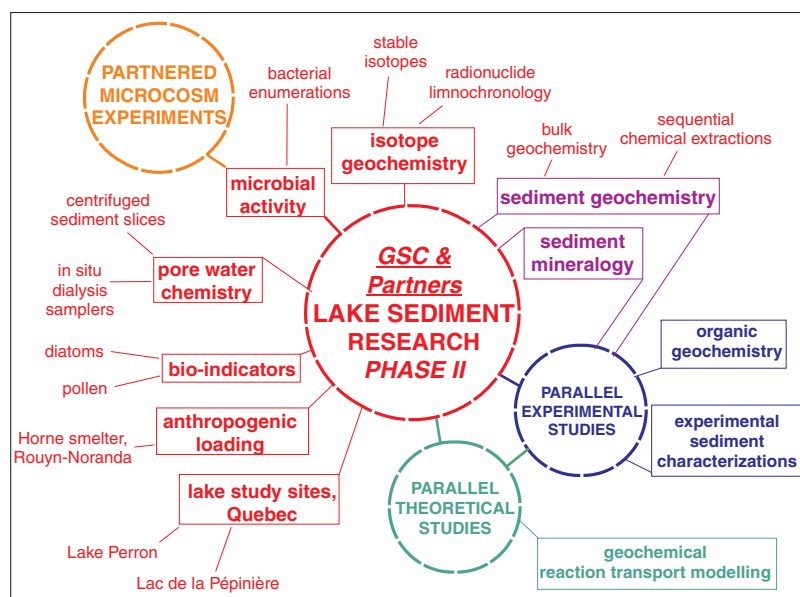


Figure 1. Project structure and measurements within GSC MITE lake-sediment research (Phase II) and partnered or parallel studies.

record the effects of natural processes that must also be documented and characterized in order to assess lake response to anthropogenic activities.

In addition to dating, stable isotope geochemistry and preserved diatom remains in lake sediments provide historical insights into lake conditions before and during industrialization. Stable isotope geochemistry and elemental ratios aid in assessing the sources of organic matter and past environmental conditions. The sulphur content and sulphur-isotope signatures in lake sediments can provide a temporal record of sulphur loading in response to smelter activity and an understanding of internal sulphur transformations (e.g. Meyers and Ishiwatari, 1995). Additionally, the historical record of lake-water acidity can be reconstructed by analyzing diatom assemblages deposited in the sediment column; diatoms are single-celled siliceous microscopic algae that serve as biological indicators of present and past lake-water conditions, including pH (Dixit et al., 1992).

The potential mobility of metals can depend on reduction-oxidation (redox) reactions during early diagenesis. Temporal shifts in the position of the redox boundary can drive a redistribution of metals in the sediment column (e.g. through changes in oxidation state and solubility followed by reductive dissolution, diffusion, and reprecipitation at another location) or through authigenic mineralization (e.g. Sholkovitz, 1985; Hamilton-Taylor and Davison, 1995). In addition to the highly reactive iron and manganese oxides and oxyhydroxides (Davison, 1985; Hamilton-Taylor and Davison, 1995), natural labile organic compounds are also likely carrier phases for metals in lacustrine sediments (Förstner, 1982; Davison, 1993); both the inorganic and organic components of the solid phase are being characterized by direct physical methods in parallel studies. Geochemical investigations also examine mineralogical controls on metal distributions in the sediment column. Various inorganic and organic sulphur

phases are being isolated and their isotopic compositions determined to understand lake response to anthropogenic sulphur input and subsequent sulphur transformations in the sediments. Stable isotopes and light-element ratios also aid in discerning the selective degradation of organic matter and element cycling within the sediment column (e.g. Meyers and Ishiwatari, 1995). In addition to measurements of bulk sediment geochemistry, quantitative analysis of metal affinities to various solid phase fractions is being tested by sequential chemical extractions of organic-rich lake sediments at two study sites. The identification and quantification of reactive solid phases and their metal loads are, therefore, being assessed by both mineralogical and geochemical analytical approaches in this study.

Diagenetic reactions within the solid phase directly affect pore-water chemistry as metals are either mobilized to the pore water or sequestered from the aqueous phase into the sediments (Sholkovitz, 1985; Hamilton-Taylor and Davison, 1995). Therefore, pore waters from the same stratigraphic intervals as the sediment slices were extracted for direct comparisons of solid- and aqueous-phase chemistry. In situ dialysis samplers (peepers; Hesslein, 1976) were also deployed by divers to sample pore water using an independent method for comparison of techniques that are expected to produce similar results (e.g. Carignan et al., 1985; Azcue and Rosa, 1996).

Diagenetic metal mobility is sensitive to redox reactions and can be modified by microbial activity (e.g. Schink and Benz, 2000). For example, increased sulphate concentrations in lakes from anthropogenic loading enhance bacterial sulphate reduction, which can lead to diagenetic sulphide formation and metal sequestration in the solid phase (e.g. Hamilton-Taylor and Davison, 1995). Bacterially mediated redox reactions (e.g. sulphate and iron reduction) can enhance the diagenetic remobilization of metals in the sediments, and are being studied in both reconnaissance microbial enumerations of lake-sediment cores and in partnered microcosm experiments that simulate lake-bottom conditions.

The resultant multidisciplinary data are intended for synthesis in developing a conceptual understanding of metal remobilization during early diagenesis for two pilot study sites. This conceptual development is essential to form a realistic geochemical reaction transport model that can be tested with the conceptual hypothesis for wider applicability to a variety of aquatic environments. Increased predictive capability is the ultimate goal of these efforts to assess metal mobility as a result of natural processes in lake sediments and in response to anthropogenic and natural perturbations in freshwater systems.

This research is possibly the first comprehensive study to integrate evidence from geochemistry, microbial populations, bio-indicators, quantitative mineralogy, and numerical

approaches to understanding the processes that control early diagenesis in a freshwater system. The results will advance interpretations of lake sediments beyond a record of simple input to incorporate the effects of early diagenesis on metal concentrations and distributions.

SELECTION OF STUDY LAKES

Two study lakes were chosen within the zone of influence of the Horne smelter, Rouyn-Noranda, from 99 lakes sampled during Phase I field studies (Kliza and Telmer, 2001). A variety of selection criteria were applied to these reconnaissance field sites to rank potential study lakes.

One of the key criteria used to verify that the study sites were within the zone of influence of the Horne smelter is evidence of increasing lake-water acidity over time using diatoms in sediment cores. As part of the regional lake studies in the Abitibi-Témiscamingue region (Fig. 2), diatom assemblages in recently deposited surface sediments (0–1 or 0–2 cm) were compared with pre-industrial layers (18–20 or 19–20 cm) to assess the qualitative change in lake-water pH over time (C. Prévost, unpub. data, 2000). Inferences of acidity were most effective in lakes with high diatom species diversity, abundance, and preservation. Sites located within the path of the major wind directions with respect to Rouyn-Noranda and with no obvious metal loading from other anthropogenic point sources (e.g. abandoned mines) further satisfied the lake selection criterion of being within the zone of influence of the Horne smelter.

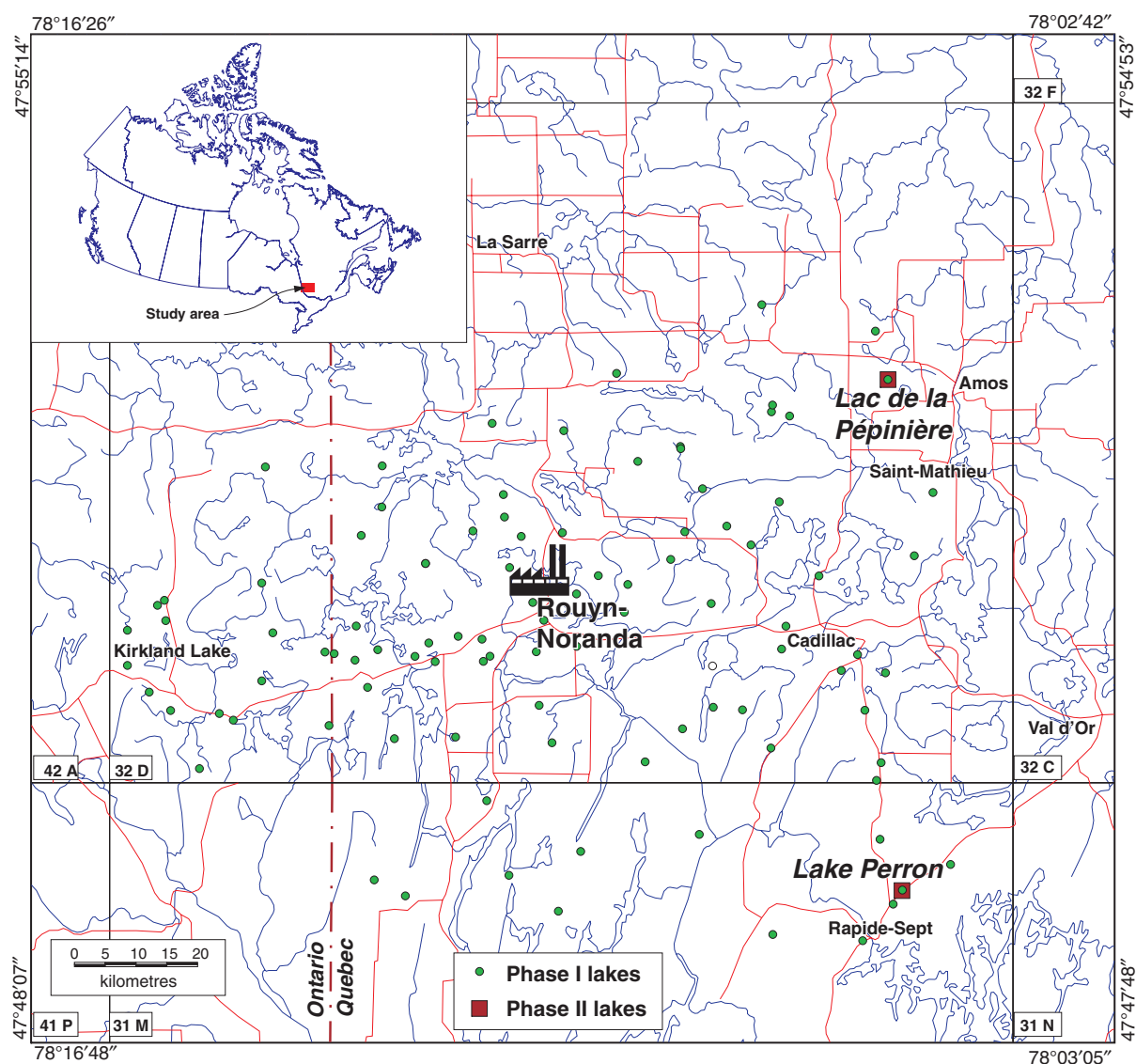


Figure 2. Location of study lakes. On the basis of diatom results available from lakes sampled in regional studies (Phase I; Kliza and Telmer, 2001), Lake Perron and Lac de la Pèpinière were selected for the second phase of research on vertical metal distributions within the sediment column.

A shift in the depth of oxygen penetration in the sediments or in the water column can cause metal remobilization as a result of changes in oxidation state and solubility (e.g. Davison, 1985). High oxygen demand under the ice cover suggests that, for most lakes of the Abitibi-Témiscamingue region, hypolimnetic anoxia develops during the winters (Wetzel, 2001); therefore, lakes with a record of complete water-column oxygenation during the summer months were considered potential study sites. Similarly, lakes with lower summer measurements of dissolved organic carbon concentrations in the water column were also given higher priority since dissolved oxygen is consumed rapidly by degradation of organic matter and can limit oxygen penetration in the sediments (Froelich et al., 1979).

An additional selection criterion was to find lakes with a high probability of undisturbed bottom sediments. Small lakes with a maximum depth between 8 and 18 m ensured a low probability of wind-driven sediment mixing and still provided adequate conditions for coring by divers and instrument deployment. Logistical considerations also necessitated choosing sites with seasonal ground access and sites that were being assessed simultaneously for parallel studies.

A set of 11 candidate study lakes emerged using the above selection criteria and existing results (*modified from Kliza and Telmer, 2001*). In a field reconnaissance of the 11 potential study sites, lake-water measurements included maximum lake water depth, pH, temperature, dissolved oxygen content, and dissolved organic carbon content, in addition to field

observations of lake accessibility and physiography. Lake Perron and Lac de la Pépinière were ranked highest as study sites that met the majority of the selection criteria (Table 1).

The two sites are likely receptors of acidic deposition from the Horne smelter with no other apparent anthropogenic sources in their vicinity. Temporal shifts in diatom assemblages suggest a history of decreasing lake-water pH at each site, despite the distances of Lake Perron and Lac de la Pépinière from the smelter (73 and 66 km, respectively). The two lakes are also currently acidic (surface water pH of 4.6 and 4.9, respectively, measured in July 2000). They are located in the path of the two prevailing wind directions recorded by weather stations at both the Rouyn-Noranda and Val-d'Or airports with major headings in the eastern quadrants (between 350°–50° and 120°–160° azimuth at Rouyn-Noranda and 330°–30° and 130°–160° azimuth at Val-d'Or airports based on hourly wind data, 1971–2001; Environment Canada, 2002a, b; Fig. 3). Lake Perron is located at an azimuth of 128° southeast of the smelter (133° southeast of the Rouyn-Noranda airport) and Lac de la Pépinière, at 55° northeast of the smelter (44° northeast of the Rouyn-Noranda airport) in the major wind directions (Fig. 2, 3). Additionally, previous regional lake surveys confirmed that metal (and metalloid; e.g. Pb, As) concentrations in Lake Perron and Lac de la Pépinière are higher in recent surface sediments (0–2 cm) in comparison with pre-industrial layers (18–20 cm; Kliza and Telmer, 2001). Although regional surveys in various environmental media suggest that the majority of

Table 1. Assessment of candidate study lakes.

Rank*	Lake	Diatom evidence	Temporal change in lake-water pH	Reconnaissance assessment
1	Lac de la Pépinière	diversity and abundance high	circumneutral-alkaline to acidic	selected lake
2	Lake Waite	striking change in diatoms (rare taxa)	alkaline to alkaline	affected by mine tailings; treated with lime**
3	Lake Perron	complete shift in dominant diatoms	acidic to more acidic	selected lake
5	Lake Hector	rich and diverse diatoms	acidic-circumneutral to acidic	inadequate ground access
6	"L7"	abundant diatoms	acidic to acidic	inadequate ground access
10	Nelson Lake	low diversity and abundance	circumneutral to weakly acidic	not in a major wind direction with respect to the smelter
15	Lake Desperiers	low diversity and abundance	acidic to acidic	dredged periodically**
18	Lake Vose	poor in diatoms	minor change in pH	inadequate ground access
***	Lake McNamara	—	—	inadequate ground access
***	Lake Lesage	—	—	inadequate ground access
***	Lake Turcotte	—	—	inadequate ground access; adjacent to acid mine drainage sites
* based on temporal changes in pH inferred from diatom analyses of recently deposited surface sediments (0–1 or 0–2 cm) in comparison with pre-industrial sediments (18–20, 19–20 cm or more; C. Prévost, unpub. data, 2000)				
** local anecdotal evidence				
*** lakes under consideration for parallel studies				

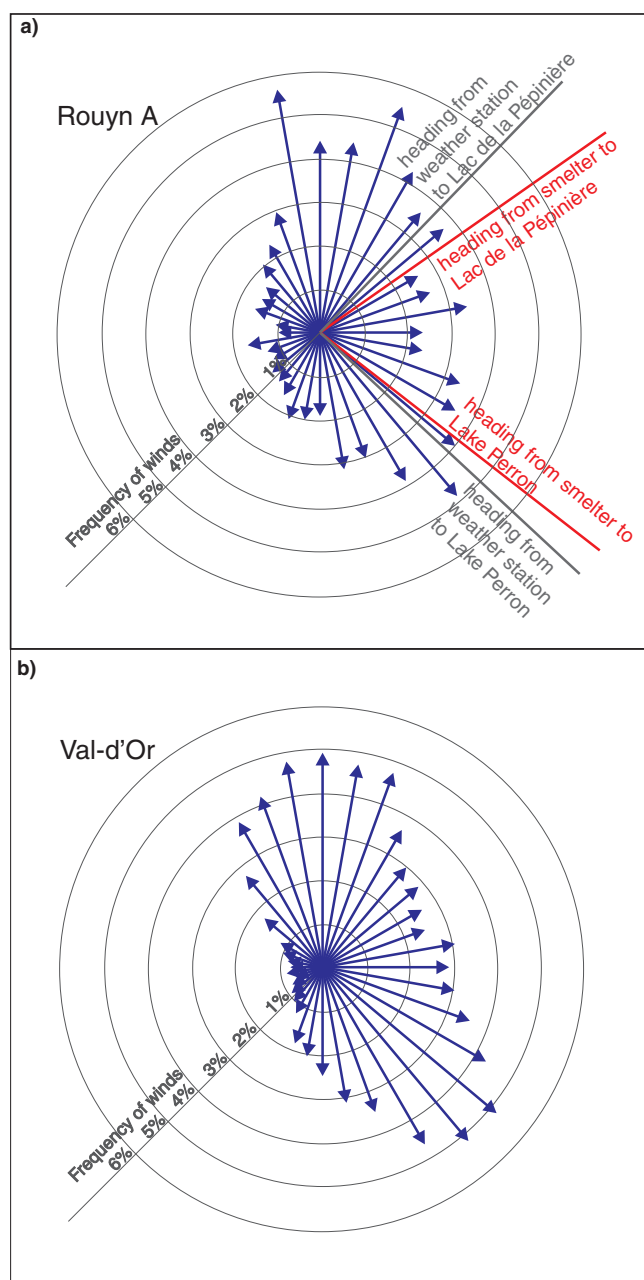


Figure 3. Wind rose diagrams. Frequency of wind direction recorded at **a)** Rouyn A (Rouyn-Noranda airport) and **b)** Val-d'Or weather stations (hourly wind data, 1971–2001; Environment Canada, 2002a, b). The orientations of the lakes with respect to the weather station (grey) and the smelter (red) are shown in a).

emissions from the Horne smelter are deposited within a 50 km radius (Bonham-Carter et al., 2005), confirmation that the two sites have been affected by the Horne smelter is emerging from geochemical and bio-indicator evidence.

Oxygen penetration into the sediments is probable in the summer since the water column profiles of Lake Perron and Lac de la Pépinière demonstrated complete oxygenation in previous samplings (August 1997; D.A. Kliza and K. Telmer,

unpub. data, 2000). Dissolved organic carbon (DOC) in surface water was lower in the two study lakes than at the other sites, <1.0 mg/L at Lake Perron and 1.4 mg/L at Lac de la Pépinière (compared with up to 6.5 mg/L at reconnaissance sites), suggesting that oxygen consumption through degradation of organic matter is more likely to occur in the sediments rather than in the water column during the summer (Froelich et al., 1979). The summer oxygen profiles and surface lake-water DOC, combined with a strong likelihood of hypolimnetic anoxia in winter, suggest a seasonal change in the position of the redox boundary.

With maximum lake-water depths of 16.5 m in Lake Perron and 12 m in Lac de la Pépinière and small lake areas and fetches, sediment mixing by wind is unlikely. No evidence of benthic fauna was found in cores retrieved from either site, indicating that sediment mixing through bioturbation or bio-irrigation is also unlikely.

Finally, seasonal ground access was adequate at the two sites with road access in summer and snowmobile access in winter. Nearby city centres (Rouyn-Noranda and Cadillac for Lake Perron and Amos for Lac de la Pépinière) also satisfied logistical requirements for field activities.

FIELD PROGRAM AND METHODS

The field program and methods included subbottom acoustic profiling during the summer of 2000 using a Knudsen 320M high-resolution echosounder, operating at 28 kHz. High quality, undisturbed sediment cores were hand-taken by divers (20–25 cm sediment penetration; Mudroch and MacKnight, 1991) so as to avoid or minimize potential disturbances, such as compaction, smearing, tilting, and mixing of small-diameter gravity cores in soft sediments with an emphasis on preserving the sediment–water interface (e.g. Baxter et al., 1981; Glew et al., 2001). The use of divers also facilitated the retrieval of large-diameter cores in unconsolidated sediments with water contents higher than 90% that could not have been recovered by coring from the lake surface. The sediments were cored at the deepest point in the profundal zone of the lakes where the depositional record is expected to be the most complete and where lake-sediment profiles have traditionally been interpreted as chronometers of metal loading (e.g. MacKnight, 1991). Cores were extruded in the field with a piston displaced by pressurized water and were sectioned vertically at 1 cm depth intervals (Mudroch and MacKnight, 1991), either under an inert atmosphere in a field glove box (Mudroch and Bourbonniere, 1991) or in the open as required for subsequent analyses. Pore water was sampled using two techniques at 1 cm depth resolution: in situ dialysis sampling and ex situ extraction of pore water from sediment slices by centrifuging (Adams, 1991; Azcue and Rosa, 1996) with sample handling under an inert atmosphere (e.g. Bufflap and Allen, 1995). In addition to water-column profiling and long-term monitoring of dissolved oxygen, pH, temperature, and conductivity (H_2O profiling system with DataSonde® III data loggers; Hydrolab® Corporation, 1997), individual lake-water samples were collected at various water depths for chemical analysis. Dissolved organic carbon was measured

by determining the total organic carbon content of water samples that were syringe-filtered through a 0.45 µm Sterivex filter cartridge. A Shimadzu 5000 Total Organic Carbon analyzer with a detection limit of 1.0 mg/L was used for the analysis. The field program included seasonal sampling during summer (June, July, and August 2000), winter (February and March 2001), and autumn (September and November 2001) with water-column monitors (DataSonde® III data loggers; Hydrolab® Corporation, 1997) deployed through a one-year cycle.

LAKE SETTINGS

Regional bedrock and surficial geology

The study sites are located within the Abitibi Greenstone Belt of northern Quebec and Ontario, which has been a major mining area of gold and base metals for nearly a century. Veillette et al. (2005) describe the bedrock geology in the area surrounding Rouyn-Noranda based on an adaptation of the litho-stratigraphic map of the Abitibi Subprovince (MERQ–OGS, 1984). A summary of the bedrock and glacial history of the Abitibi-Témiscamingue region is also provided by Veillette et al., (2005). An extensive clay plain and well developed esker systems are the main surficial features of the Abitibi glaciated terrain. The absence of bedrock outcrops and the deep depressions formed by kettle lakes suggest that the glaciofluvial sediments are probably several tens of metres thick in places.

The Abitibi-Témiscamingue region is characterized by extensive and thick glaciofluvial and glaciolacustrine deposits that cover the bedrock over large areas. The major eskers of central Abitibi result from subglacial meltwaters that flowed in conduits under high hydrostatic pressure and produced the central coarse-textured core typical of Abitibi eskers (Veillette, 1996; Veillette et al., 2005). The abrupt transition from high energy flow in tunnels within the glacier to unconstrained conditions in the lake at the ice margin produced the observed sequence of overlapping subaqueous fans characteristic of the Abitibi eskers. This process accounts for the exceptional volume and width of the eskers, which do not have the sharp crests typical of eskers deposited in subaerial environments. Chains of kettles and kettle lakes aligned parallel to the longitudinal axis of the eskers mark the position of the coarser materials within the esker core. With the retreat of the ice sheet, Glacial Lake Ojibway waters covered the eskers, modified their surfaces, and locally even buried them under thick layers of varved clay. Wave action from the gradual lowering of Glacial Lake Ojibway further modified the surface of the eskers by displacing large volumes of sand toward the esker flanks. The combination of these processes produced landforms of large proportions with a central higher topography of coarser granular materials flanked by thick, predominantly sandy deposits having poorly defined contacts with the surrounding glaciolacustrine deposits. These large bodies of granular deposits are confined on each side by the impervious clay layers deposited in the deepest parts of Glacial Lake Ojibway and locally are aquifers of economic importance.

Lake Perron and Lac de la Pépinière are both kettle lakes that formed within thick glaciofluvial deposits. Kettles generally occur as steep-sided depressions that result from melting of ice blocks detached from the ice margin and subsequently buried in the sediments left behind by the retreating glacier. The melting of the buried ice blocks can occur over a time scale of several hundred years.

Lake Perron surficial geology

Lake Perron is the larger of the two kettle lakes. It is located in the largest body of granular deposits of the Abitibi-Témiscamingue region, the Harricana Moraine, which reaches a width of approximately 7 km in the vicinity of Lake Perron (Fig. 4; Veillette, 1987). The sedimentology of this interlobate moraine is similar to that of an esker. It consists entirely of stratified sand and gravel, the surface of which has been reworked extensively by wave and/or wind action. East of Lake Perron, postglacial winds produced a spectacular field of large parabolic dunes. Their positions on the east flank of the moraine and their orientations indicate past prevailing wind directions to the east and southeast. The dunes were later stabilized by peat development.

The Lake Perron subbottom acoustic profiles indicate three acoustic facies (Fig. 5). The lowermost unit does not show any internal structure, indicating the presence of acoustic basement below which acoustic energy is prevented from further penetration. The acoustic basement likely corresponds to compact glaciofluvial sediments. This unit is typically observed at lake margins and occasionally at shoals where postglacial sediments have not been deposited. The middle unit is up to 6 m thick and exhibits strong, coherent, parallel internal reflections that are closely spaced. The reflections become more closely spaced as the sediments drape over the underlying structure and are more widely spaced in the deepest water. This unit represents glaciolacustrine sediments deposited after, and possibly during, decomposition of the ice core during the formation of the kettle lake. The type of sediment depends largely on water depth and proximity of the sediment supply at the time of deposition. In most areas of the lake where water depths exceed 5 m, the lake bed is covered by an acoustically transparent layer with no visible internal structure. It is thickest in the depressions on the lake bottom and likely represents the gyttja or dy that was cored from the deepest point in the lake (Fig. 6). Gyttja and dy are soft, hydrous, freshwater sediments composed of the remains of particulate organic matter in addition to allochthonous and autochthonous inorganic components; gyttja contains less than 50% organic matter and dy, more than 50% (Wetzel, 2001).

Lac de la Pépinière surficial geology

Lac de la Pépinière is a smaller and shallower kettle lake in the Saint-Mathieu-Berry esker, which is the aquifer from which the city of Amos draws its water supply (Fig. 7; Veillette et al., 2003). Farther south along the esker, east of the village of Saint-Mathieu (shown in Fig. 2), another major aquifer will be used to produce bottled water on a commercial

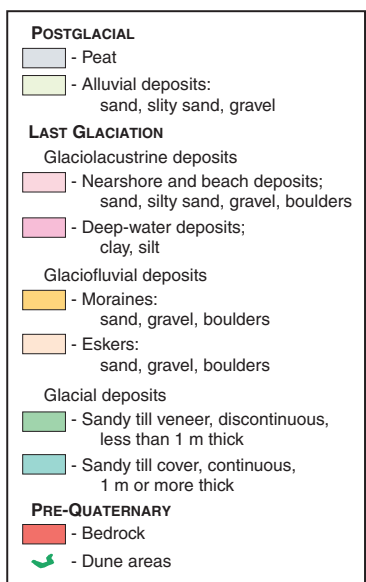
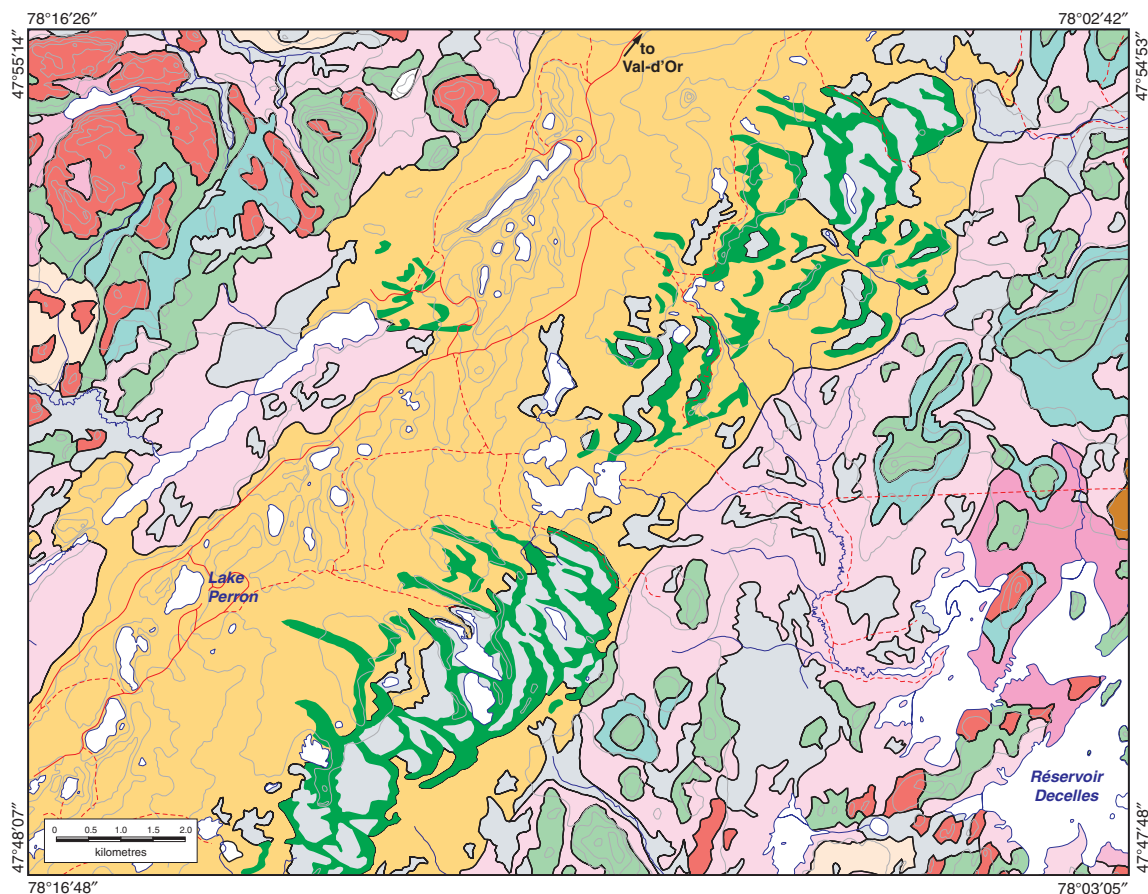


Figure 4.
Local surficial geology of Lake Perron (after Veillette, 1987)

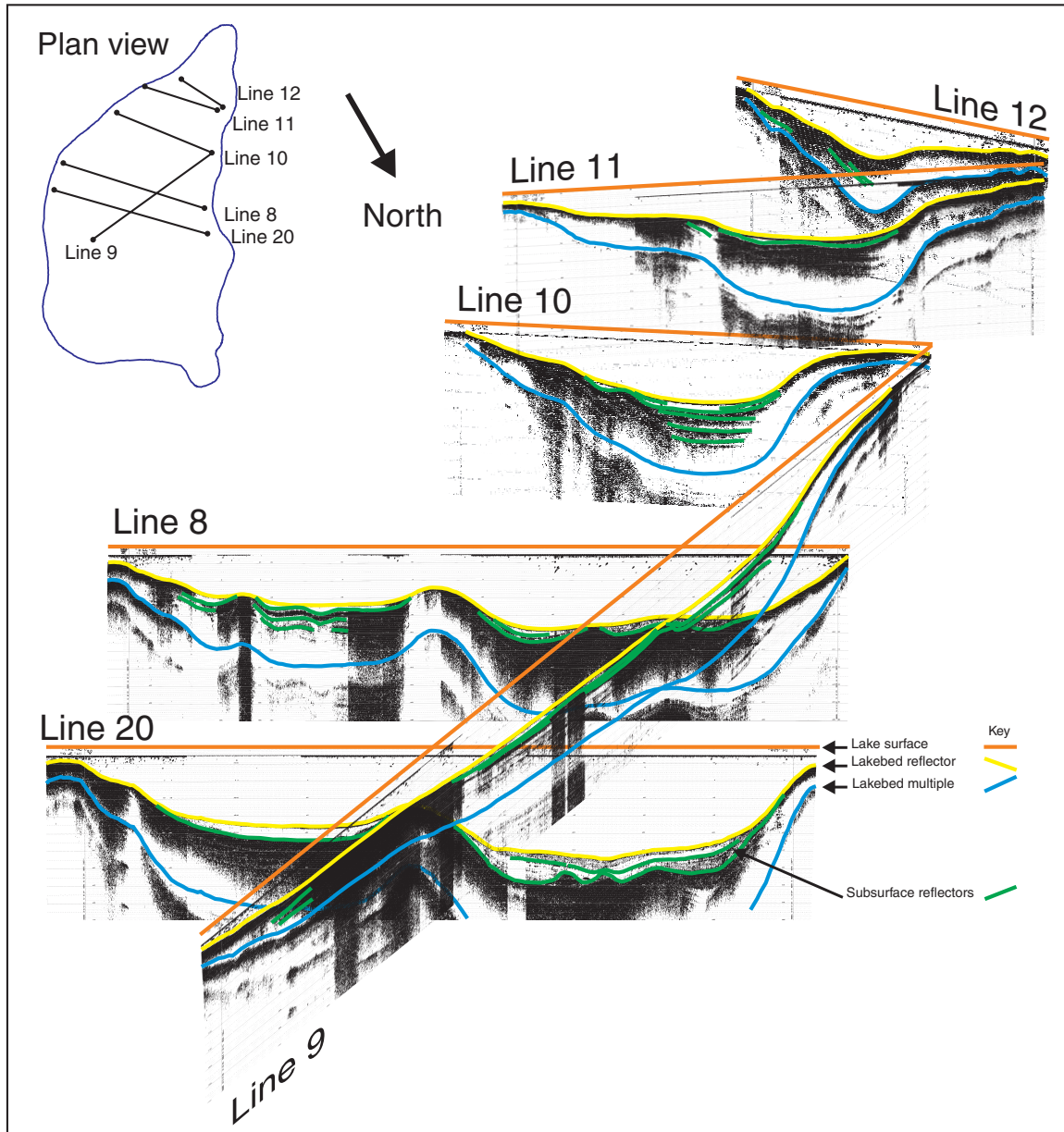


Figure 5. Subbottom acoustic profiles of Lake Perron with sonar transects (inset); bathymetry given in Figure 6.

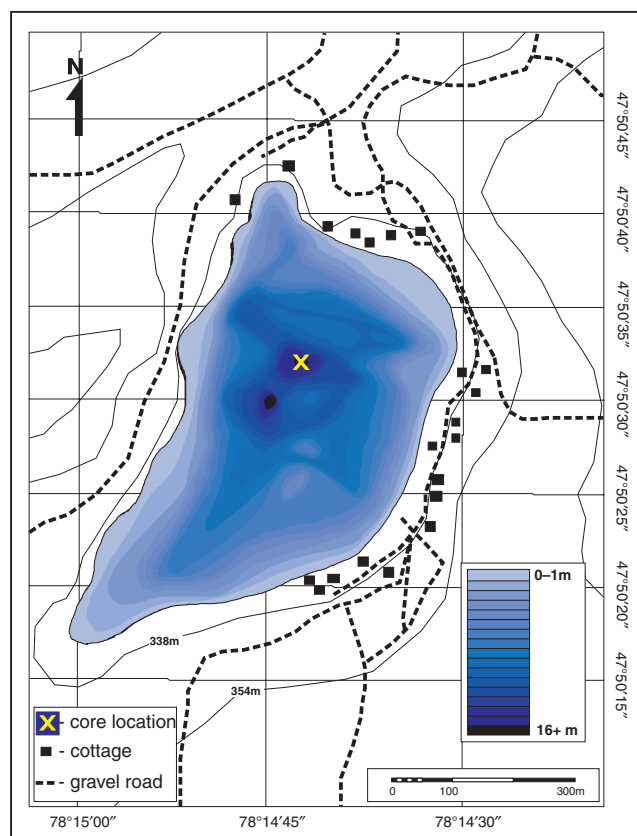


Figure 6. Bathymetry of Lake Perron.

basis. Numerous springs are found along the Saint-Mathieu-Berry esker. These can emerge at the interface between clay and granular deposits.

The acoustic subbottom profile of Lac de la Pépinière reveals two acoustic facies (Fig. 8). The lowermost one is a hard impenetrable reflector, likely composed of compact glaciofluvial sediments. Overlying this acoustic basement is a poorly stratified reflector that probably represents organic-rich sediments. This layer is up to 2 m thick, except near the shore, where it thins. Interestingly, the poorly stratified layer appears to be absent at the deepest point in the lake (Fig. 9), suggesting that the lake bottom is composed of the lowermost clastic unit. However, gyttja or dy is present in the profundal zone of the lake and was cored, but it could not be distinguished from the underlying sediments, probably due to the lack of sufficient acoustic contrast between the water column and the gyttja or dy. A gain setting that is appropriate for imaging clastic sediments may not necessarily detect the sediment–water interface, particularly when the upper sediments are as hydrous as gyttja or dy.

Study lake characteristics

The characteristics of Lake Perron and Lac de la Pépinière are summarized in Table 2. Since both are perched kettle lakes without any significant streams, their major hydrological inputs are from overland flow directly into the lakes and direct precipitation. Groundwater seepage to the study lakes

is unlikely. In the Abitibi-Témiscamingue region, lake-water temperature and pH distinguish groundwater within the esker aquifers from lake water in perched kettles that are sealed off from the regional water table by fine organic matter (J.J. Veillette, pers. comm., 2004). Perched kettle lakes with an impervious layer of fine organics at the base and without any major inflows or outflows have high summer water temperatures (20°C) and low pH (4–5). In contrast, springs from the esker aquifers are colder (6°C and 9°C) and have higher pH (6.5–8.5) than lake waters. The high summer water temperatures of Lake Perron and Lac de la Pépinière, combined with their acidities (pH of 4.6 and 4.9, respectively), suggest that groundwater input to the lakes is not significant. Lake Perron and Lac de la Pépinière are located at the boundary between two vegetation zones within the boreal forest of the Abitibi-Témiscamingue region, with dominantly yellow birch to the south and white birch to the north (Richard, 1988). Mixed forests dominated by deciduous trees are prevalent within the two lake watersheds.

Lake Perron

The watershed that contains Lake Perron is mostly deciduous although it is a mixed forest containing white birch, silver birch, black spruce, and red spruce with 55 to 95 years of growth to a height of 12 to 17 m (Ministère de l'Énergie et des Ressources du Québec, 1986a, b). The area adjacent to the southwest shoreline of Lake Perron is wet and marshy and largely barren of trees. The adjacent watersheds are also predominantly deciduous mixed forests with white birch, silver birch, black spruce, red spruce, poplar groves, and jack pine. Clear-cut patches from 1978, 1979, and 1987 have been documented in the area with reforestation that has allowed up to 20 years of growth; the surrounding forest is up to 95 years old. Localized epidemic disease has been reported but only as mild cases. The history of forest fires near Lake Perron suggests that three events have occurred within a 25 km radius since 1945 (A. Belleau, pers. comm., 2002; Table 3).

Approximately 20 summer cottages are found along the northeast perimeter of Lake Perron; they are accessed by a gravel road. A few attempts were made to stock the lake with trout during the 1950s; these were unsuccessful, presumably due to a lack of tolerance for lake-water acidity (P. Bérubé, pers. comm., 2001). Current residents use two sources of water for their domestic supply: groundwater from a communal well located approximately 50 m north of the lake edge and lake water. The lake water was remarkably clear with an unusually deep photic zone that allowed for the prolific growth of aquatic macrophytes on the lake bottom at water depths of up to 16.5 m.

The lack of summer thermal stratification in Lake Perron is striking (Fig. 10). Despite its maximum depth of 16.5 m, fetch of 830 m, and surface area of 24 ha, the water column remains well mixed and isothermal throughout the summer months (approximately 19°C measured in July 2000). However, given the lake's geometry and morphometry, the water column is expected to achieve stable stratification at or near the time of maximum heat content in the summer (e.g. Gorham and Boyce, 1989). Wind-induced mixing of the

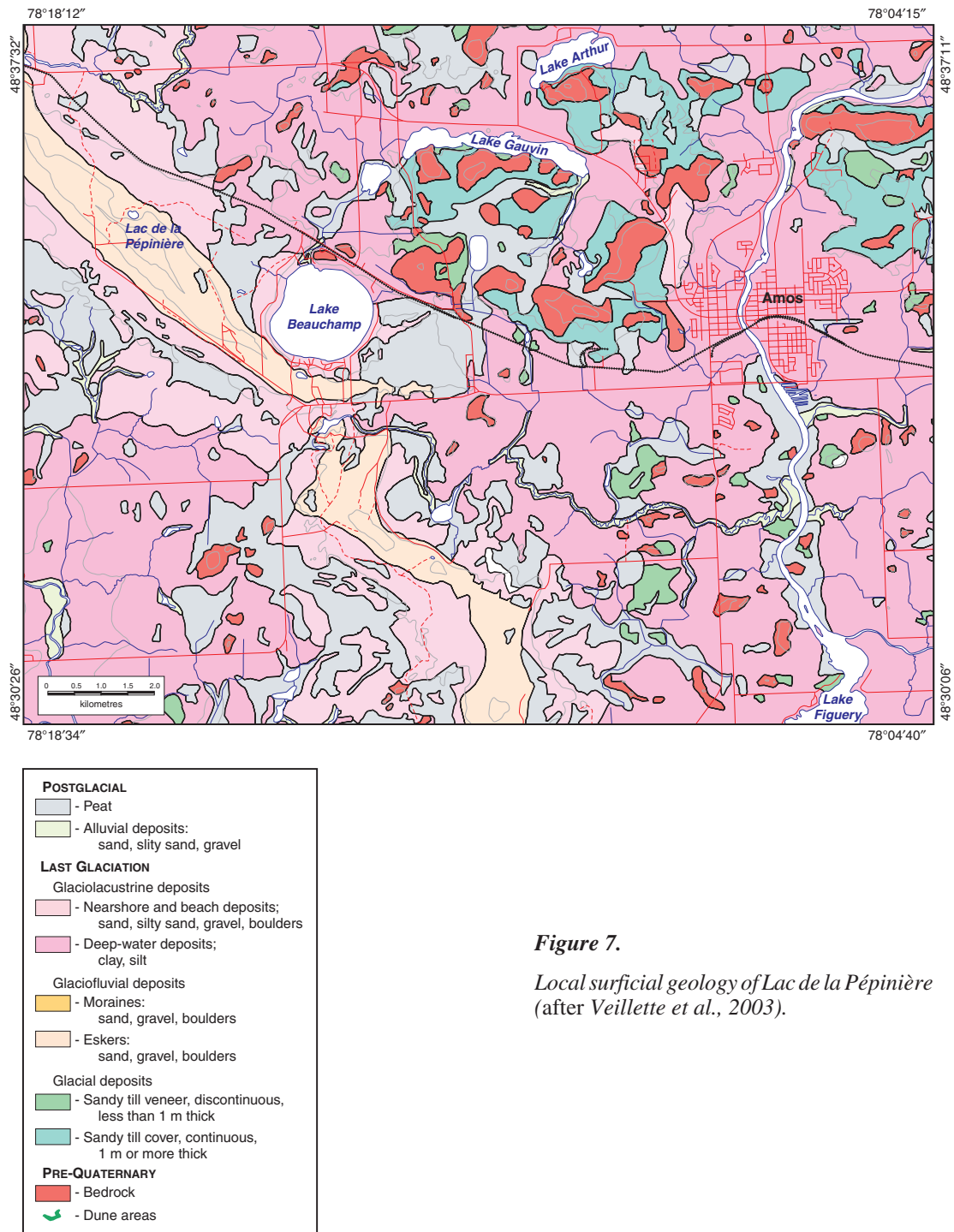


Figure 7.
Local surficial geology of Lac de la Pépinière
(after Veillette et al., 2003).

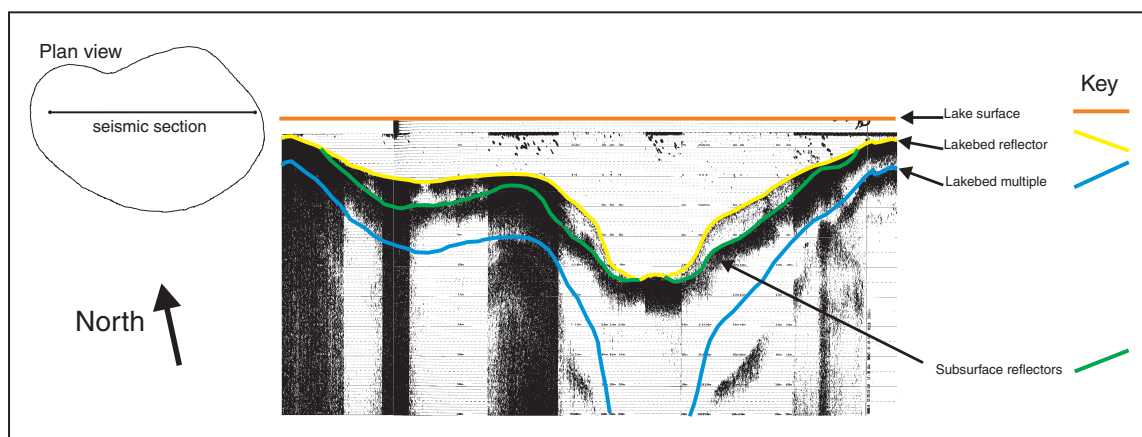


Figure 8. Subbottom acoustic profile of Lac de la Pépinière with sonar transect (inset); bathymetry given in Figure 9.

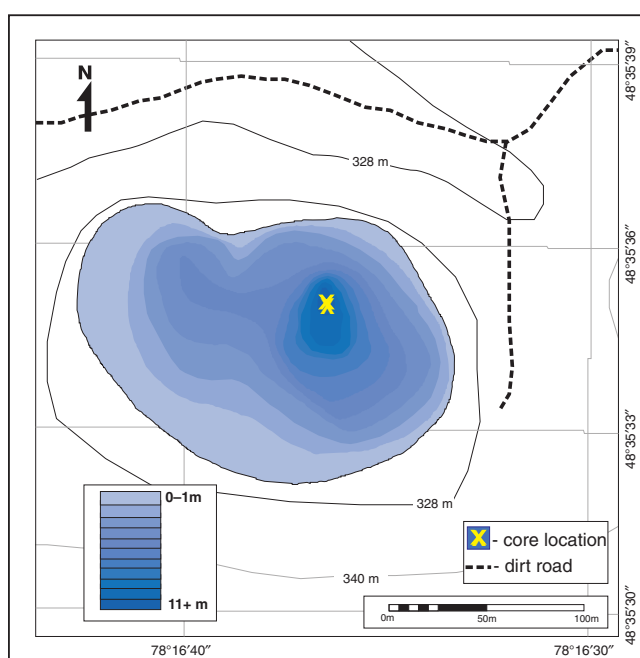


Figure 9. Bathymetry of Lac de la Pépinière.

water column may provide enough energy to either destroy the existing stratification or prevent it from forming and stabilizing after spring melt. Long-term lake-water data recorders have been deployed to investigate the temporal thermal dynamics of the water column and to assess when and if stratification occurs. Inverse thermal stratification that developed under the winter ice cover was as expected (Fig. 10; Wetzel, 2001).

Profiles of dissolved oxygen concentration in the water column of Lake Perron demonstrated a strong probability of oxygen penetration into the sediments in summer with a completely oxygenated water column (near 100% saturation). In contrast, oxygen depletion in the deep water column is likely during winter ice cover with a drop from near saturation at 1 m below the surface to 53% saturation at a depth of 13 m

(Fig. 10). In order to avoid disturbing the surface sediments, the descending profiling system was not allowed to reach the depth of the sediment–water interface at 16.5 m.

The sediments at Lake Perron were greenish-brown gyttja or dy with little variability in texture downcore, although the spatial distribution of aquatic macrophytes was heterogeneous within the deepest part of the lake basin. During the 2001 winter sampling, one unused grouping of cores contained patches of macrophytes that were either at surface and cored to a depth of 20 to 25 cm, or buried to a depth of 20 cm under the overlying gyttja or dy.

Lac de la Pépinière

The immediate surroundings of Lac de la Pépinière appear to be largely second-growth forest and include a provincial forestry centre. The local watershed and adjacent basins contain white birch, silver birch, black spruce, red spruce, fir, and jack pine with a canopy height of between 12 and 22 m and a forest age up to 85 years (Ministère des Ressources naturelles du Québec, 1995). South of Lac de la Pépinière, forests were cut to develop a series of gravel pits along the Saint-Mathieu-Berry esker. Because of the permeability of the esker deposits, the watersheds in the area generally have good drainage and remain dry at surface. There have also been three forest fires within a 9 km radius of the lake, all occurring in 1953 (A. Belleau, pers. comm., 2002; Table 3). One of the fire sites extended right up to the lake's western shoreline.

Recreational swimmers, users of all-terrain vehicles, and members of a snowmobile club frequent Lac de la Pépinière, also known locally as 'Lac Dalton' (F. Talbot, pers. comm., 2001). Summer access to the lake is by a dirt road that also connects to the nearby gravel pits within the Saint-Mathieu-Berry esker. Lac de la Pépinière is located in a pronounced depression within the esker; evidence of erosion along its banks suggests that surficial spring runoff could be a substantial input to the lake.

Table 2. Summary of lake settings.

	Lake Perron	Lac de la Pépinière
location	N47°50'29.4", W78°14'45.4"	N48°35'35.3", W78°16'36.5"
*change in lake-water pH	acidic to more acidic	circumneutral-alkaline to acidic
surface lake-water pH, July 2000	4.6	4.9
orientation from the smelter; azimuth	SE; 128°	NE; 55°
distance from the smelter (km)	73	66
dissolved organic carbon (mg/L) in surface lake water, July 2000	≤1.0	1.4
maximum lake-water depth (m)	16.5	12
bioturbation	low probability	low probability
physiography; surficial geology	kettle lake; Harricana Moraine	kettle lake; Saint-Mathieu-Berry esker
bedrock geology	acid intrusive rocks	mixed volcanic rocks
inputs	runoff, precipitation	runoff, precipitation
area of lake (ha)	24	2
fetch (m)	830	200
vegetation	dominantly deciduous mixed boreal forest	dominantly deciduous mixed boreal forest including a provincial forestry centre
current land use	~20 summer cottages along NE lake perimeter; recreational swimmers, users of all-terrain vehicles, snowmobilers	recreational swimmers, users of all-terrain vehicles, snowmobilers
water column, July 2000	well mixed; clear	stratified
** aquatic biota	no fish or benthic fauna; water snakes, frogs, beavers	no fish or benthic fauna; water striders, beavers
<p>* based on temporal changes in pH inferred from diatom analyses of recently deposited surface sediments (0–1 or 0–2 cm) in comparison with pre-industrial sediments (18–30, 19–20 cm or more; C. Prévost, unpub. data, 2000)</p> <p>** based on field observations and anecdotal evidence from local residents</p>		

Table 3. Summary of forest fires in the vicinity of the study sites.

	Year	Distance and heading from the lake
Lake Perron	1972	8.5 km; W 270°
	1953	15.2 km; NW 280°
	1951	24.8 km; NE 84°
Lac de la Pépinière	1953	at lake edge; western lake shoreline
	1953	8.5 km; NW 296°
	1953	8.7 km; SE 130°
A. Belleau, pers. comm., 2002		

The water column of Lac de la Pépinière is stratified during the summer with a thermocline depth of 9.5 m (Fig. 11). The fact that the lake stratifies is consistent with lake geometry (fetch of 200 m, surface area of 2 ha), but the thermocline depth is twice that expected for a lake with a maximum water depth of 12 m in middle North America (Gorham and Boyce, 1989) despite the expected interannual variability (Wetzel, 2001). The lake may have adjusted to an imposed wind stress by increasing the depth of the thermocline

while preserving stratification of the water column. Inverse stratification has developed under the ice cover in winter, suggesting that Lac de la Pépinière is dimictic (Wetzel, 2001).

During the summer, the water column was also completely oxygenated, suggesting that the redox boundary penetrated the sediments (Fig. 11). Under winter ice cover, however, the concentration of dissolved oxygen decreased with water depth from a maximum of 72% saturation at 1 m below surface to 7% at 2 m above the sediment–water interface. Deeper measurements were not taken so as to preserve the integrity of the sediment–water interface. The hypolimnion likely becomes anoxic in winter and the position of the redox boundary likely shifts seasonally from within the sediments in summer to the water column in winter. Given the high organic content of the bottom sediments, the sediment oxygen demand can offer considerable stress to the hypolimnetic oxygen content to a state of total depletion.

Dark brown organic-rich gyttja or dy was present at the bottom of Lac de la Pépinière with a slight decrease in moisture content and an increase in compaction at a sediment depth of 12 to 14 cm. Pine needles were encountered at

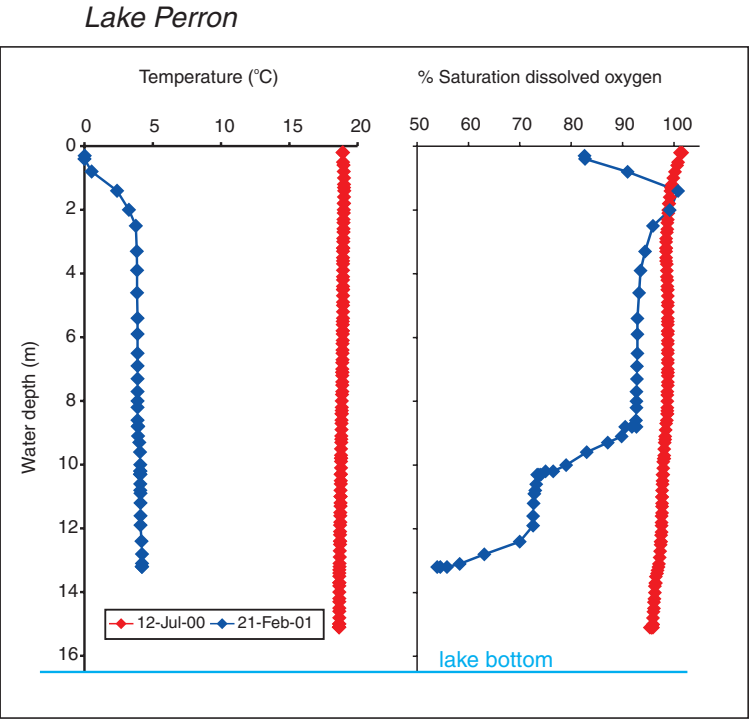


Figure 10.
Seasonal water-column measurements of temperature and dissolved oxygen concentrations at Lake Perron.

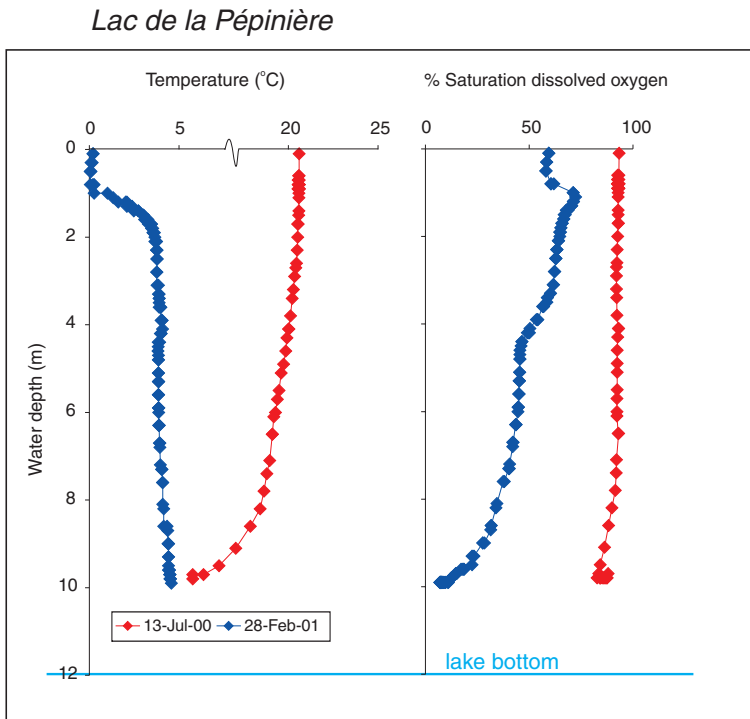


Figure 11.
Seasonal water-column measurements of temperature and dissolved oxygen concentrations at Lac de la P  pini  re.

various depths within the core during summer 2000 sampling, whereas heterogeneously spaced discrete woody fragments were found during both summer and winter sampling seasons. Trees close to the shoreline could have shed their needles into the lake or fallen in leaving behind the pine needles and patchy wood fragments during early decomposition. A visual inspection of erosive features along the shoreline also suggests that spring runoff from the immediate surficial drainage basin could transport a considerable organic load, including pine needles and wood fragments, to the lake.

During sampling dives, divers noted that when the core liners were carefully inserted into the bottom sediments of both study sites, the entire surface of the lake bottom surrounding the sampling site would shake because of the gelatinous nature of the gyttja or dy. However, preliminary analytical results (e.g. sediment dating; A.S. Dixit, S. Alpay, S.S. Dixit, and J.P. Smol, unpub. rept., 2004) indicate that the sediments have remained undisturbed by potential physical mixing processes such as bioturbation, groundwater seepage, and wind mixing or by coring disturbances despite the high water contents (>90%) of the surface sediments.

SUMMARY AND FUTURE DIRECTIONS

Preliminary observations suggest that the two study sites are suitable to address the original aims of this study using a novel interdisciplinary synthesis of results from diverse fields within the Earth sciences. Vertical metal mobility within the sediment column is possible in part because of the seasonal shift in the redox boundary that causes changes in chemical speciation and solubility. The two lakes are likely within the zone of influence of the Horne smelter in Rouyn-Noranda, on the basis of evidence from diatom assemblages in modern and pre-industrial sediments, lake orientations in the major wind directions with respect to the smelter, and the absence of any other apparent anthropogenic metal source in their vicinities. The two lakes chosen are geochemically distinct with different histories of acidification, different geometries, and different orientations with respect to the main point source.

As results continue to be generated, interpreted, and integrated, an increased understanding of metal cycling and diagenetic metal remobilization in lake sediments is developing. Furthermore, new hypotheses and quantitative information that result from the interdisciplinary approach taken will enable the development of a conceptual understanding of metal mobility during early diagenesis that can provide practical predictive capability for use by government and industry. Applications include water-resource management, environmental impact assessments, risk assessment and risk management with respect to bioavailability of potentially toxic metals, predicting sediment response to environmental perturbations (e.g. anthropogenic loading, physical disturbance), and interpreting industrial and pre-industrial sedimentary records of metal loading.

This paper has presented the rationale for questioning the use of vertical metal distributions in lake sediments as historical archives of chronological metal loading in light of potential metal mobility during early diagenesis. The objectives of the project can be met through the strength of a multidisciplinary approach and interdisciplinary interpretations. Unlike other lake-sediment studies, an effort has been made to establish and explicitly describe the criteria and steps for the selection of study sites, given the benefit of previous extensive regional studies (Kliza and Telmer, 2001). Finally, the settings of the two acidic kettle lakes, Lake Perron and Lac de la Pépinière, have been characterized in order to place ongoing and future studies within a contextual framework.

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