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Disruption of the geochemical metal cycle during mining: Multiple isotope studies of lake sediments from Schefferville, subarctic Québec

Stéphane Aebischer [a], Christophe Cloquet [b,*], Jean Carignan [†], Charles Maurice [c], Reinhard Pienitz [a]

[a] Takuvik, UMI 3376 CNRS-ULaval et CEN (Centre d'Etudes Nordiques), Laboratoire de Paléoécologie Aquatique, Université Laval, Québec, QC G1V 0A6, Canada
[b] CRPG, UMR 7358 CNRS/université de Lorraine, 15 rue notre dame des pauvres BP20, 54501 Vandoeuvre les Nancy, France
[c] Bureau de la connaissance géoscientifique du Québec, Ministère de l'énergie et des Ressources Naturelles, Canada

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ABSTRACT

Iron mining in Schefferville (1939–1977) in subarctic Quebec has left behind large numbers and quantities of tailing deposits. The impact of past mining activity on aquatic ecosystems in the Schefferville area has been studied using geochemical and isotopic (Pb, Zn and Fe) analyses of lake sediments, ore deposits, tailings and epiphytic lichens. Analysis of two sediment cores from Lakes Dauriat and Oksana reveals that the surface geochemical cycle of the Schefferville area has been profoundly disturbed by anthropogenic activity such as mining. Disturbances were particularly abrupt at the transitions from pre-mining to mining and mining to post-mining periods. Elemental and isotopic analyses of the lake sediments reveal four different end-member contributions to the lake sedimentation, with changes in terms of sources and source contribution observed throughout the sedimentation history. End-members were identified using Pb, Zn and Fe isotopes and are consistent for each element. Lead isotope ratios vary from highly radiogenic ($^{206}Pb/^{204}Pb = 27$) to less radiogenic ($^{206}Pb/^{204}Pb = 17.7$) from the bottom to the top of the sediment cores. Iron isotope compositions vary from −0.2‰ to 1‰, the latter value remaining constant throughout the sedimentary history of Lake Oksana. A systematic difference in the Zn isotope ratios of the two lakes is also observed, and can be explained by local differences in basin lithology. In order to identify pollution sources, samples from ores and tailings and epiphytic lichens were measured as proxies of mining activity, lithology, and atmospheric deposition, respectively.
The impact of anthropogenic activity is clearly evident in the sediment records and results from mining activity, as well as local urban and industrial activities (waste water inputs). Long-range atmospheric deposition also accounts for some of the variations in isotopic composition measured in the sediments. The systematic coupling of Pb isotopes with Zn or Fe isotopes allows us to identify and constrain the metal sources that contributed to sediment contamination. This study demonstrates that disturbance due to mining activities is very effective in rapidly modifying lake sediment composition. Though the disturbances remained local in terms of geographic extent, the damage to the aquatic ecosystem has been significant and may persist for decades.

1. Introduction

Mining operations produce negative impacts on the environment through the generation of mineral and other wastes that are transported across the land and through the atmosphere. This waste includes products used for ore extraction, as well as waste rock and anthropogenic waste associated with mining activities. The environmental impacts associated with mining mostly result from mechanical damage to soils, ecosystems and landscapes and from the effects of acid mine drainage (Dudka and Adriano, 1997). Large amounts of dust are emitted into the atmosphere during ore extraction, pre-treatment (crushing) and transport, especially in open pits, which generate significantly more atmospheric pollution than underground mines (Dudka and Adriano, 1997). Ore-processing by smelting and refining releases many trace elements in particulate and gaseous forms (Dudka et al., 1995; Simonetti et al., 2004). These particles are later deposited in terrestrial and aquatic environments via dry and wet deposition (Baron et al., 2006; Sivry et al, 2008). In aquatic ecosystems, trace elements released by mining activity are found in both water and sediments. The concentrations of chemical elements in sediments usually increase significantly at the beginning of mining activity and then, as leaching of metals into aquatic systems is a relatively slow process, peak in the post-mining period (Dudka and Adriano, 1997).

Prior to the mining, the Schefferville area was occupied by nomadic people (Humphrys, 2013) such as the Montagnais and the Naskapis (Barbeau, 1987). The first mining company was officially launched in 1947, for Fe ore extraction, but mining exploration is known to have started during the early 20th century. The town of Schefferville was

* Corresponding author.
E-mail address: cloquet@crpg.cnrs-nancy.fr (C. Cloquet).
† Deceased.

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built during the same period, in parallel with the development of the mines. To transport and market the ore, a railway was constructed, connecting Schefferville with Sept-îles (Humphrys, 2013). Schefferville experienced almost exponential growth up until the beginning of the 1980s. Wastewater was drained into Lake Dauriat, located in the town centre, until 1975, when a wastewater treatment plant was built. However, fierce competition from mining companies in Brazil and Australia, led to a decline in ore mining and growth of the city was halted. The Schefferville mines eventually closed in November 1982 (Barbeau, 1987). However, due to a relatively recent inflation in the price of iron, mining activity around Schefferville restarted in June 2011.

The mining of metals has the potential to disturb the surface geochemical cycle of metals, resulting in significant increases in the concentrations of trace and toxic elements in the environment (Thornton, 1996). Metal concentrations and isotopic compositions analysis of archives are very useful for obtaining information on trace element contamination. Lake sediments are particularly useful records of changes in metal inputs to the environment (Gallon et al., 2005; Thapalia et al., 2010), being formed through a combination of atmospheric deposition, deposition from the watershed's drainage system and precipitation of organic and mineral matter from the water column. Epiphytic lichens provide a proxy for atmospheric deposition alone, and can therefore complement sediment records for source identification.

The use of Pb isotopes for identifying sources of contamination has become routine in the last few decades (Graney et al., 1995; Weiss and Shotyk, 1999; Simonetti et al., 2000; Monna et al., 2004; Cloquet et al., 2015). This is largely because Pb isotopes are not altered by chemical or physical processes (Ault et al., 1970). Lead does not fractionate and therefore retains the isotopic composition of its source. Identification of past Pb source contamination has been studied in sediments (Nedjai et al., 2011; Thevenon et al., 2011) and peat bogs (Weiss and Shotyk, 1999; Baron et al., 2005). The use of epiphytic lichens in trees has also proved very effective for identifying long-range atmospheric transport (Carignan and Gariépy, 1995; Carignan et al., 2002).

Zinc (Zn) and Iron (Fe) isotopic compositions can be used in addition to Pb because, unlike Pb, these can be modified by a range of different processes. With the advent of Multicollector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS), the analytical precision of Zn (Albarede, 2004; Cloquet et al., 2008) and Fe (Dauphas and Rouxel, 2006) isotope ratio measurements has become smaller than the natural variations. This has opened up new opportunities for the study of the biogeochemical cycling. Zn emitted into the atmosphere through smelting and refining is enriched in the lighter Zn isotopes, whereas Zn in tailings and slags is enriched in the heavier Zn isotopes (Cloquet et al., 2006b; Dolgopolova et al., 2006; Sivry et al., 2008; Sonke et al., 2008; Mattielli et al., 2009). Distinctly light δ^{66}Zn values have been measured in urban runoff (Cloquet et al., 2006b; Chen et al., 2008; Chen et al., 2009). The identification of different mining periods has been achieved using Zn isotopes (Borrok et al., 2009; Thapalia et al., 2010), demonstrating that Zn isotopes can be used as tracers of environmental contamination. In addition, iron is the sixth most abundant element in the universe and represents 35% of the Earth's mass. It is also the main ore present in the Schefferville area. These three isotopic systems are therefore complementary and can provide different types of information on pollution sources.

In this study, we report the down-core changes in the geochemical and isotopic (Pb, Zn and Fe) composition in two sediment cores from two lakes in the Schefferville area (Lake Dauriat and Lake Oksana). Lake Oksana was chosen specifically because of its remoteness from the mining area in that it has therefore recorded the integrated signal of atmospheric pollution (as opposed to proximal mining activities), 2) Lake Dauriat was selected because of its proximity to the town of Schefferville. This lake therefore provides an integrated signal of the impacts of different types of urban pollution (gas, sewage, municipal waste etc.). In order to determine the chemical and isotopic compositions of the different metal sources in the Schefferville area lakes, geochemical and isotopic analyses were performed on epiphytic lichens (current atmospheric signal of the study area), as well as on ore deposits and tailings (anthropogenic signals related to the past Fe mining).

A combination of climatic change (i.e., warming of arctic regions and melting of permafrost) and growing economic pressure (i.e., Quebec's *Plan Nord*), has made areas like the Schefferville region increasingly accessible and attractive to mining companies. The main objective of this research is to gain a better understanding of the multiple disturbances that have affected the dynamics and kinetics of the surficial geochemical cycles of metals associated with mining activities in northern landscapes like the Schefferville area.

2. Study site

Lakes Dauriat (54°48′17.52″N, 66°49′27.31″W) and Oksana (54°47′35.85″N, 66°47′16.05″W) are located near the town of Schefferville (Fig. 1), in the New Québec Orogen (NQO), also known as the Labrador Trough. The NQO is located on the border of the Superior and Churchill geological provinces (Clark and Wares, 2004). It is composed of rocks of Lower Proterozoic age (ca 2.2 to 1.8 Ga) comprising three sedimentary facies (two of which are volcanic in origin and the other metamorphic in origin). These sequences contain orthoquartzites, dolomites, iron formations, greywackes and conglomerates of sedimentary rocks (Dimorth, 1981). The sedimentary rocks of the Schefferville area were deposited in marine and fluvial environments (Clark and Wares, 2004). The area lies partly within the Sokoman formation and partly within the Ruth formation. During the end of the Lower Proterozoic, the Sokoman formation and underlying rocks were faulted and folded. The rocks of the Schefferville area are mainly composed of quartz, iron oxides and carbonates that have been chemically precipitated. During its formation, the ore from the Schefferville area was a taconitic-poor iron ore (25–30% Fe). During the Mesozoic, the experienced meteoric leaching resulted in a decrease in Si/Fe and Si/Mn, thereby making the ore exploitable (60% Fe; Leslie and Bourque, 1984). The Schefferville iron ore is composed of two main types of deposit: the ore call “soft ore” and the “rubble ore”. The “soft ore” mainly consists of hydroxides (limonite and goethite). The “rubble ore” is mainly formed from/of goethite originating from the fragmentation and cementation of the “soft ore” (Leslie and Bourque, 1984).

The Schefferville area is located in a forest-tundra vegetation zone (Plan Nord, 2011) mainly composed of lichens (*Cladonia*), dwarf birch (*Betula nana*), shrubs of the genera *Kalmia* and *Vaccinium* and crowberry (*Empetrum nigrum*), as well as black spruce (*Picea mariana*) and white spruce (*Picea glauca*) trees. The catchments of lakes and rivers are mostly occupied by larch (*Larix laricina*) (Journaux and Taillefer, 1957).

The climate of the region is subarctic, characterised by long and cold winters and short but relatively warm summers (Government of Quebec, 2010). Between June and August, the three warmest months, the average monthly temperature is around 10 °C. Near the town of Schefferville, the prevailing wind direction is northwest with an average speed >15 km/h throughout the year (Environment Canada, 2011).

3. Materials and methods

Two sediment cores from Lakes Oksana and Dauriat were initially drilled in 1999. The two lakes were sampled again in March 2012 in order to complete the sediment chronology of the last fifteen years. Sediment cores were collected with a short Aquatic Research Instruments percussion corer (40 cm length), in roughly the middle of the lakes. The water column measures 3.5 m at Lake Dauriat and 12 m at Lake Oksana. Lake Dauriat has one tributary (south of the lake) and one effluent (north-west of the lake). The core was collected from outside the draining network. Lake Oksana is a “Kettle lake” i.e., has no tributary or effluent. Cores were mainly composed of thin and sandy sediments and organic matter. The colour of the Lake Dauriat sediment changed from brown to black along the length of the core, reflecting the change