



Original Articles

Atmospheric dispersion of trace metals between two smelters: An approach coupling lead, strontium and osmium isotopes from bioindicators



David Widory*, Geneviève Vautour, André Poirier

Geotop-UQAM Case postale 888, Succursale Centre-Ville, Montreal, H3C 3P8, QC, Canada

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ABSTRACT

Bioindicators, by naturally cumulating the impacts of chemical contaminants over time, have demonstrated their added value in evaluating environmental quality. We studied Pb, Sr and Os isotope systematics from *Cladonia rangiferina* lichens collected along a transect between two smelting complexes located in Rouyn-Noranda and Sudbury (Canada) to identify sources of these metals in the Abitibi region and to delineate the extent of their respective emission plumes. Results show that metals present in the study area are explained by the contamination of the regional background by deposition from the atmospheric emissions of the different smelters over distances up to 250 km. A rough estimate of the respective metal contributions of each smelter to the lichen samples was calculated. At low metal concentrations, lichen samples indicate that dispersion plumes may differ for the Pb and Os contamination, perhaps to be linked to differential volatility.

1. Introduction

Mining activities have been recorded as early as 500 BCE when Egyptians sank mineshafts in search for gold (Notton, 1974). Later, during the Copper/Bronze Age and the Roman Empire, mining continued releasing metals into the environment (Rauch, 2010). Mining in Canada has evolved from a Native American exploitation activity in the 1800's into an economic development lever between 1900 and 1960 (Hipwell et al., 2002). While within the last 50 years environmental concerns began shaping the mining industry there is still a need for monitoring and precisely characterising industrial activities in mining regions to establish metal background levels in the environment and to uncover potential zones of metal contamination (Lapalme, 2003). Mining in the Abitibi Gold Belt (Canada) started with the 1909 gold rush (Dixon, 1979). The Archean Abitibi greenstone belt forms the local bedrock and is mainly composed of old compressed and metamorphosed volcano-sedimentary rocks (Dimroth et al., 1984; Daigneault et al., 1990). The Horne copper smelter, located in Rouyn-Noranda (Quebec province) near the Ontario border, is one of the main active smelters in the region. The smelter processes local and worldwide ores. It also is the largest recycling facility for used electronic and computer parts in North America (Gallon et al., 2006). Previous studies have shown that the particles rejected into the atmosphere by the Horne smelter greatly influence the overall lead (Pb) budget of local and regional aerosols, sediments and flora (Simonetti et al., 2000; Carignan et al., 2002; Simonetti et al., 2004). Two other active smelters, Vale

Inco Copper Cliff and Sudbury Integrated Nickel Operation (Glencore), are located within the same geographical area ca. 300 km, in Sudbury (Ontario). The Vale Inco plant smelts nickel-copper bulk concentrates from various sources and the Glencore nickel-copper concentrates from Sudbury, Raglan and XNA (Australia) ores. The Glencore smelter also processes custom-feed materials in the form of concentrates and secondary products.

Environmental monitoring has been widely based on the study of elemental concentrations around mining activities to identify contamination resulting from atmospheric transport and deposition (e.g. Telmer et al., 2006). Recent studies however demonstrated that a multi-isotope approach could unambiguously backtrack sources of air pollutants and help characterise secondary processes occurring post-emission (e.g. Widory et al., 2004; Geagea et al., 2007). The isotope tracking of atmospheric pollutants usually involves direct sampling of both gases and filtered aerosols (e.g. Monna et al., 1997; Veron et al., 1999; Harrison, 2012), or indirect sampling of meteoric water or snow (e.g. Nakano and Tanaka, 1997). However, the use of bioindicators to monitor/assess the quality of different environments offers numerous advantages, such as the possibility of bio-concentrating both organic and inorganic contaminants and of accessing to time-integrated information (Conti and Cecchetti, 2001). Mosses and epiphytic lichens are widely recognised as reliable bioindicators for evaluating air quality due to their ability to accumulate chemical elements in amounts usually exceeding their physiological needs (Nimis et al., 2002). This attribute is due to physiological properties such as a lack of cuticle, the absence

* Corresponding author.

E-mail address: widory.david@uqam.ca (D. Widory).

of protective organs (which could limit the adsorption of toxic substances) and a large exchange surface. This is also why they are capable of absorbing both soluble and insoluble mineral nutrients; they survive by maximizing their uptake of atmospheric aerosols and precipitations while minimizing loss (Gerdol et al., 2002). Limitation of the biomonitoring approach is that it yields a time-averaged value over the lifetime of the bioindicator that can be difficult to assess (Sutton et al., 2004).

Due to the toxic nature of the complexes it forms Pb adversely impacts human health, especially children who present greater intake rates (Yoshinaga et al., 2012). Pb stable isotopes (^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb) have thus been widely studied for several purposes, including monitoring Pb accumulation in sediments (e.g. Gobeil et al., 1995; Poirier, 2006) and to trace its origins (e.g. Simonetti et al., 2000; Carignan et al., 2002), including in bioindicators (e.g. LeGalley et al., 2013). Studies are based on the possibility of discriminating the main atmospheric sources of Pb by their specific Pb isotope ratios. For example, in eastern North America the $^{206}\text{Pb}/^{207}\text{Pb}$ ratios of most of the major Pb vectors are well defined: i) Natural Pb (taken from sediments younger than 1802) vary from 1.210 to 1.216 (Gallon et al., 2005), ii) the average Canadian aerosols vary between 1.167 and 1.177 (Simonetti et al., 2000, 2004), iii) the average US aerosols vary between 1.173 and 1.195 (Simonetti et al., 2000; Bollhöfer and Rosman, 2001), iv) leaded gasoline which dominated emissions from about 1930 to 1990 in North America shows values from 1.15 to 1.183 (Gallon et al., 2005), and v) local Abitibi ores between 0.92 and 1.04 (Deloule et al., 1989; Franklin, 1983).

Strontium (Sr) isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) have also been used in environmental studies for tracing sources of aerosols (e.g. Geagea et al., 2007; Widory et al., 2010). Sr isotopes can be useful biological markers as there is no fractionation among soil, plants and herbivores (Flockhart et al., 2015). Natural sources have a large range of Sr isotope ratios, from 0.715 to 0.725 (or more) for granites, 0.707–0.709 for carbonates, 0.703 for fresh basalts and 0.7092 in the ocean (e.g. McArthur et al., 2001). $^{87}\text{Sr}/^{86}\text{Sr}$ ratios measured in atmospheric emissions from the main industrial sources also greatly vary: steel plants and chemical waste incinerators (0.7091 and 0.7087, respectively; Geagea et al., 2007), coal combustion and cement plant (0.709–0.710 and 0.7095–0.7175, respectively; Widory et al., 2010). Aerosols from the US have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging from 0.70819 to 0.71266 (Simonetti et al., 2000).

Osmium (Os), due to the harmful and volatile nature of its tetroxide (OsO_4) coupled to its increased usage or presence in various industrial products, including automobile (car catalytic converters) and mining, has also recently been studied for environmental issues (e.g. Poirier and Gariépy, 2005; Rodushkin et al., 2007). Os is a trace metal from the platinum-group elements (PGE) commonly found as an impurity among the other PGEs because of its strong association with them (Poirier and Gariépy, 2005). Rodushkin et al. (2007), coupling Os concentrations and their corresponding isotope ratios in bioindicators collected in Sweden, characterised the dispersion of the contamination plume from a nickel smelter up to 100 km away from it. In the Abitibi region (Canada) Os isotopes were mostly studied around the Sudbury region to characterize the $^{187}\text{Os}/^{188}\text{Os}$ ratios of local ores and host rocks (Morgan et al., 2002). The Sudbury basin formed following a meteorite impact about 1,8 Gy ago (Davis, 2008). It displays a crustal Os isotope signature ($^{187}\text{Os}/^{188}\text{Os} = 0.90\text{--}1.17$), (Morgan et al., 2002), consistent with impact-induced melting (Dickin et al., 1996). The Abitibi mining region extends across the Quebec-Ontario border and is formed by two major mining districts, the Abitibi gold belt (Au-Cu) from Wawa (Ontario) to Val d'Or (Quebec) and the copper-nickel (Cu-Ni) belt in Sudbury, Ontario. To our knowledge Os isotope ratios have not been published yet for this mining region.

Here we present Pb, Sr and Os concentrations and stable isotope ratios from bioindicators (i.e. lichens) collected along a transect between Rouyn-Noranda and Sudbury with the aim of characterising the

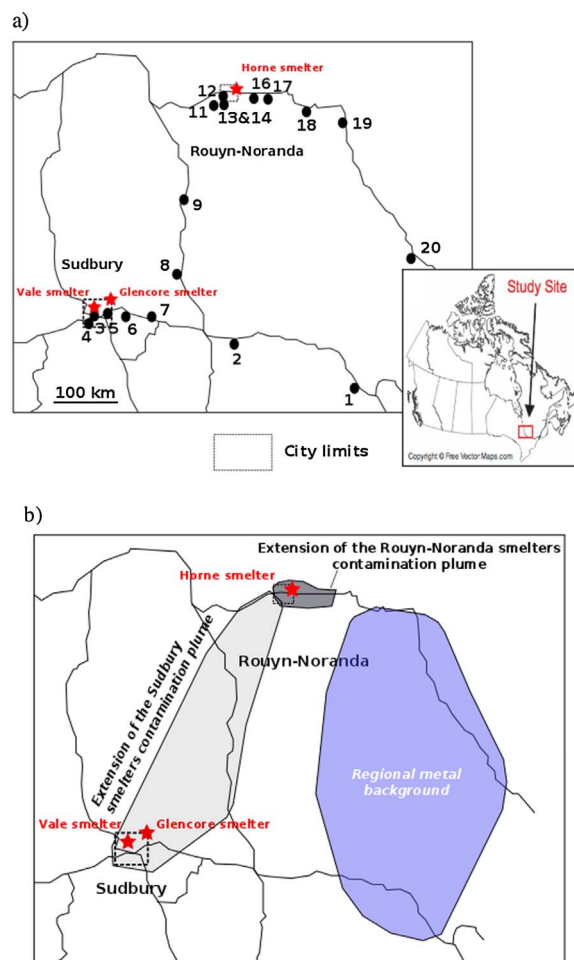


Fig. 1. A) Lichen samples locations in the Abitibi region. Numbers refer to sample numbers in Table 1. B) An assessment, based on the study of the Pb and Os isotope systematics, of the extension of the smelters contamination plume as well as of the regional background is shown. Samples 2 and 7 that are giving distinct conclusions are excluded from the assessment.

extent to which the two smelting complexes are impacting their surrounding environment.

2. Materials and methods

Eighteen *Cladonia rangiferina* lichen (also known as reindeer or caribou lichen) samples were collected (Fig. 1a) during the fall of 2014 along a transect between the two active smelters located 300 km apart in the cities of Sudbury (Ontario) and Rouyn-Noranda (Quebec). Five samples (1, 2 and 18–20) were collected far from the smelters (> 100 km) and not under dominant winds coming from them in order to try to characterise the regional background levels for Pb, Sr and Os. The *Cladonia rangiferina* is a terricolous (epigenous) lichen that was selected as i) it is widely spread in the study region and ii) soil contamination was minimised by taking the lobe tips as samples.

Lichens, not bigger than 5 cm in diameter or roughly younger than 10 years old (assuming a growth rate of $0.4\text{--}0.5\text{ mm year}^{-1}$; Scotter, 1963; Pegau, 1968), were sampled in cleaned plastic bags. For each sampling location at least 5 distinct lichen samples were collected. Samples were taken away from roads, in parks or hiking trails when possible. Lichen samples were first washed with ultrapure Milli-Q water, dried at $50\text{ }^\circ\text{C}$ for 24–48 h. Lobe tips were collected from the thallus and then crushed using an agate mortar. Because they potentially represent the main local sources of industrial contamination, three samples corresponding to the main ores processed in the Abitibi

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