The role of pollution versus natural geological sources for lead enrichment in recent lake sediments and surface forest soils

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Abstract This paper assesses the role of airborne pollution and natural geological sources for lead enrichment in lake sediments and in surface soils of boreal forests. This assessment is based on analyses of stable lead isotopes ($^{206}$Pb and $^{207}$Pb) and lead concentrations in sediment cores (>30 lakes), ombrotrophic peat and soil samples in Sweden. The $^{206}$Pb/$^{207}$Pb ratio and concentration profiles in the sediment cores change synchronously over the last 3,000 years in different lakes, temporal concentration changes in sediments and peat deposits are very consistent, and these temporal concentration changes coincide well with the history of lead production in Europe. The $^{206}$Pb/$^{207}$Pb ratio is almost the same in all soil mor samples (1.152±0.009; $n$=94), and similar to values recorded in aerosols, despite very high and different $^{206}$Pb/$^{207}$Pb ratio of the mineral soil in the C-horizon (1.3–1.7). This study provides evidence that lead enrichment in recent sediments and peat, and in the mor layer are caused by deposition of pollution lead and not natural processes.

Keywords Forest soils · Lake sediments · Lead enrichment · Natural geological sources · Pollution

Introduction

There is still debate over the importance of atmospheric deposition in comparison to natural geological sources for the contribution of trace metals in rural and remote ecosystems. For example, Rasmussen (1998) is a strong advocate for the significance of natural sources. In a review that examines the methods of estimating the relative contribution of natural and anthropogenic sources to trace metals in the environment, Rasmussen (1998) writes: “A large number of studies use changes in the metal content of natural samples of varying age (e.g., organic lake sediments, organic forest soils) to infer changes in atmospheric input to the ecosystems where these samples are collected. Unless such studies address the accumulation processes which cause metals to become naturally enriched in surface soil and organic sediments, it is difficult to verify their conclusions. Uncertainty in the interpretation of metal enrichment in surface forest soils as a historic record of pollution arises from the fact that some naturally occurring metals such as lead typically become concentrated in the upper few centimeters of undisturbed soils, due to their incorporation in living plants and accumulation in decomposing remains in the humus layer”.

Rasmussen (1998) argues for the need for a ‘geoscience perspective’ to distinguish between natural and anthropogenic sources of heavy metals to the environment. We address the issue of natural versus anthropogenic lead and Rasmussen’s concern for erroneous interpretation of lead pollution history from lake sediments and forest soils. The basis for the assessment is an extensive data-set comprising analyses of lead concentrations and stable lead isotopes of >1,400 samples of sediment, peat and forest soil. In addition to this use of different environmental archives, a main asset in our studies has been the analysis of stable lead isotopes ($^{206}$Pb/$^{207}$Pb ratios). Lead isotope analysis is a powerful tool to distinguish between lead from different sources, in this case anthropogenic and natural sources (Ault and others 1970; Shirahata and others 1980; Rabinowitz 1995; Véron and Church 1997).

It has proven to be particularly useful in the Swedish environment because of the very large difference in lead isotope ratios between uncontaminated soils and sediments, on the one hand, and ancient to Medieval airborne lead pollution, as well as modern airborne lead pollution, on the other (Fig. 1).
The $^{206}$Pb/$^{207}$Pb ratios in old unpolluted lake sediments (>3,000 years old), mineral soils (>70 cm depth; Brännvall and others 1997, 1999, 2001a, 2001b; Bindler and others 1999), ores from Roman/Medieval times (Gröger and others 1966; Brill and Wampler 1967; Wedepohl and others 1978; Sugden and others 1993; Stos-Gale and others 1995; Rolh 1996; Rosman and others 1997) and modern aerosols in Sweden (Hopper and others 1991)

The dataset we use for this assessment comprises:

1. Sediment cores from >30 lakes cored between 1986 and 1999 with the aim of studying temporal and geographic trends in the atmospheric deposition of lead pollution (Renberg and others 1994, 2000; Brännvall and others 1997, 1999, 2001a). The main criteria for site selection in these studies were to obtain a reasonable geographic coverage of Sweden and to choose lakes with a minimum of land-use disturbance. No particular consideration was taken to the mineralogy of the catchment soils. Therefore, the data-set gives a random, average picture over a large geographic area.

2. Sediment cores from Lake Storasjö, which is situated in an area with anomalously high lead concentration in bedrock and soils. Till samples from a 40-km$^2$ large area around Storasjö have concentrations between 60 and 300 µg g$^{-1}$ (Sundblad 1997). This lake was specifically selected for this study to compare and contrast the lead concentrations and isotope ratio profiles in the sediment from this lake with profiles from the other lakes, which are surrounded by mineral soils with more normal, low lead concentrations (<18 µg g$^{-1}$, Melkerud and others 1992).

3. Peat from three ombrotrophic bogs in southern Sweden (Brännvall and others 1997; Bindler and others 1999). Ombrotrophic bogs receive pollutants only from the atmosphere (Gore 1983) and hence should give a reliable record of pollution history, unless metal migration occurs in the peat.

4. Soil profiles (up to 60–110 cm in depth) from eight sites (11 profiles) with old-growth boreal forests (Bindler and others 1999; Brännvall and others 2001b) and samples of the mor layer (the organic surface layer) from the forest near Storasjö. Besides showing lead concentration and isotope ratio changes with depth, this data-set gives an opportunity to compare lead concentrations and lead isotope signatures in mor samples from areas with very different parent soil materials in order to assess the influence of natural and pollution lead in surface forest soils.

The study sites and the lead analyses

Figure 2 shows the location of the study sites. In this paper we only show data from 11 lakes, namely those in which entire core sequences covering the period from the present-day and >3,500 years back in time were analysed.