



Sources, transport and sinks of beryllium in a coastal landscape affected by acidic soils

Mats E. Åström^{a,*}, Changxun Yu^a, Pasi Peltola^a, Jason K. Reynolds^d,
Peter Österholm^b, Miriam I. Nystrand^b, Anna Augustsson^a, Joonas J. Virtasalo^c,
Linda Nordmyr^b, Antti E.K. Ojala^c

^a Department of Biology and Environmental Science, Linnaeus University, SE-39182 Kalmar, Sweden

^b Department of Geology, Abo Akademi University, FI-20500 Åbo, Finland

^c Geological Survey of Finland (GTK), PO Box 96, FI-02151 Espoo, Finland

^d School of Science and Health, Western Sydney University, Sydney, Australia

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Abstract

Beryllium (Be) sources, transport and sinks were studied in a coastal landscape where acidic soils (acid sulfate soils) have developed after drainage of fine-grained sulfide-bearing sediments. The study included the determination of total abundance and speciation of Be in a variety of solid and aqueous materials in both the terrestrial and estuarine parts of the landscape. A major feature was that despite normal (background) Be concentration in the sulfide-bearing sediments, the Be leaching from these sediments after O₂-exposure and acid sulfate soil development were extensive, with concentrations up to 76 µg L⁻¹ in soil water, 39 µg L⁻¹ in runoff and 12 µg L⁻¹ in low-order streams. These high Be concentrations were mainly in the solution form (i.e., passing a 1 kilodalton filter) and modelled to be dominated by free Be²⁺. The extensive Be release within, and leaching from the acid sulfate soils was controlled by pH, with a critical value of 4.0 below which the Be concentrations increased strongly. Although plagioclase and mica were most likely the main carriers of Be within these soils, it is suggested that other minerals such as Be hydroxides, Al hydroxides carrying Be, and Be sulfides are the main contributors of the abundance of dissolved Be in the acidic waters. When the acidic and Be-rich creek water was neutralized in the estuary of relatively low salinity, the dominating solution form of Be was removed by transformation to particles, reflected in the suspended particulate matter that had hydroxylamine hydrochloride extractable Be up to 17 mg kg⁻¹ and ammonium acetate EDTA extractable Be up to 4 mg kg⁻¹. In corresponding pristine materials (parent material of the acid sulfate soils) in the catchment, the median Be extractability with these reagents were only 0.3 and 0.05 mg kg⁻¹, respectively. As the Be-rich suspended particulate matter ultimately became benthic sediment, the Be was preserved in terms of total concentrations but underwent to some extent changes in speciation, including release from hydroxides and concomitant scavenging by organic matter and particle surfaces. © 2018 Elsevier Ltd. All rights reserved.

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1. INTRODUCTION

Beryllium (Be), recognized as a highly toxic metal (Taylor et al., 2003; Shah et al., 2016), generally occurs in

low concentrations in natural surface waters. For example, the concentrations (<0.45 µm fraction) in Eastern UK rivers are generally less than 0.1 µg L⁻¹ with a mean of 0.02 µg L⁻¹ (Neal, 2003), in North American rivers below 0.013 µg L⁻¹ (Kusakabe et al., 1991), in 807 streams across Europe generally less than 0.1 µg L⁻¹ with a median of 0.009 µg L⁻¹ (Salminen et al., 2005), and in 1344 streams

* Corresponding author.

E-mail address: mats.astrom@lnu.se (M.E. Åström).

in the Eastern Barents region (the taiga and tundra areas in Finland and NW Russia) below the quantification limit ($0.005 \mu\text{g L}^{-1}$) in more than 70% of the water samples (Salminen, 2004). The Be concentrations can, however, be substantially elevated, such as in Eastern UK rivers where the concentrations occasionally can reach as high as $29 \mu\text{g L}^{-1}$ despite near-neutral pH conditions (Neal, 2003).

Enhanced mobilization of Be is typically associated with low-pH conditions and acidification. This is indicated by inverse correlations between pH and Be concentrations in several surface-water datasets (Vesely et al., 1989; Neal, 2003), an increase in Be concentrations in stream water artificially acidified for a short period (Norton et al., 2000), elevated Be concentrations in surface waters as a result of acid-precipitation effects on soils and weathered rock (Kram et al., 1998; Navratil et al., 2002), large Be mobilization during acidification in catchments where the rocks are enriched in Be and contain the mineral beryl (Brown et al., 1992; Navratil et al., 2002; Neal, 2003), and detectable losses of Be from the top soil during natural and anthropogenic acidification (Navratil et al., 2002). These features and trends are supported by experimental data showing a substantial decrease in Be sorption on clay minerals and biotite with decreasing pH (You et al., 1989; Aldahan et al., 1999; Boschi and Willenbring, 2016a), and a substantial increase in the proportion of dissolved Be with decreasing pH in kaolinite or silica systems both in the absence and presence of humic acid (Takahashi et al., 1999).

On many coastal plains an acidic soil, referred to as “acid sulfate soil”, develops when the groundwater table drops as a result of natural or artificial drainage in fine-grained calcite-poor sediments (mostly Holocene) containing pyrite and/or metastable Fe sulfides (Dent and Pons, 1995). In Europe, the largest occurrences of these soils are found on the coastal plains east of the Gulf of Bothnia (the northernmost part of the Baltic Sea). In this area (hereafter referred to as “Ostrobothnia”), multi-element hydro-geochemical studies have identified a correlation between Be concentrations in stream water and the areal percentage of fine-grained (clay-silt) sediments ($r_{\text{Pearson}} = 0.85$) (Astrom and Bjorklund, 1995) and arable land ($r_{\text{Spearman}} = 0.68$) (Roos and Astrom, 2005). The correlation with arable land is not explained by addition of Be via agricultural activities, because the chemicals and fertilizers applied on the arable land here are not expected to contain Be. Instead, these correlations are most likely explained by leaching of Be from acid sulfate soils developed on the fine-grained sediments that are typically used as farmland (arable land) in these areas (Yli-Halla et al., 1999). Additionally, the three streams with the highest Be concentrations (0.45 , 0.79 and $1.17 \mu\text{g L}^{-1}$) of all those streams (1344) included in the geochemical atlas of the Eastern Barents region are located to Ostrobothnia (Salminen, 2004). Taken together, these data and features suggest that acid sulfate soils supply significant amounts of Be to surface waters, in a similar manner as they do for several other metals (e.g. Al, Mn, Ni, Co, Zn, rare earth elements) in a variety of climatic zones (Minh et al., 1997; Sundstrom et al., 2002; Macdonald et al., 2007; Welch et al., 2009).

Knowledge of Be in acid sulfate soil settings is lacking, and hence a potentially important element in terms of Be leaching to aqueous systems in coastal regions has not been considered properly. In order to fill this knowledge gap, we carried out a comprehensive field-based study in an acid sulfate soil landscape in Ostrobothnia. The overall aim was to define, and determine the underlying mechanisms of the release, transport and sinks of Be in landscapes where acid sulfate soils are widespread and a significant source of acidification and metal mobilization (Sundstrom et al., 2002; Boman et al., 2010).

2. SETTING

The Vörå creek (hereafter “the creek”) and its estuary, located in a rural area with no Be point sources in Ostrobothnia, are the main focus of this study. This creek has a catchment area of 223 km^2 of which approximately 20% is covered by acid sulfate soil (farmland), 15% by peat/histosols (mainly mires) and the rest by other soil types including podzols (mainly forests) (Astrom et al., 2010). The acid sulfate soils in this region are typically 1–2 m deep and developed on Holocene fine-grained (silt-fraction dominating) sediments that were deposited in brackish marine waters and are now distributed on coastal plains as a result of the postglacial isostatic land uplift. These sediments have an aluminosilicate matrix and contain 0.2–2% sulfur in the form of pyrite and metastable iron sulfide (Astrom and Bjorklund, 1997; Boman et al., 2008). Although a small proportion of the acid sulfate soils may have developed due to natural drainage, the vast majority have developed as a result of artificial farmland drainage since the 1960s. No lake basins exist in the catchment. The estuary of the creek is generally shallow (<17 m deep), non-tidal and well-sheltered from wave action by surrounding land area and islands. The annual mean temperature is $3 \text{ }^\circ\text{C}$, the mean annual precipitation is approximately 500 mm, and from December to April there is snow and ice in this area.

3. METHODS

3.1. Samples

The study utilizes a variety of samples collected in previous sampling campaigns and described in detail elsewhere, as summarized in Table 1.

Samples of acid sulfate soils were collected at 10 sites (Fig. 1) and include bulk samples of the acidic soil horizon and of the underlying near-neutral sulfidic fine-grained sediment. The former is referred to as “acid sulfate soil” and the latter as “parent material”.

Waters from the acid sulfate soil fields include (i) soil water from the unsaturated zone collected from two lysimeters on four occasions, (ii) runoff collected from a confined plot on 26 occasions, and (iii) stream water collected from 7 low-order streams on one occasion (Fig. 1). These water samples were filtered through $0.45 \mu\text{m}$ membranes. Groundwater was collected from the parent material with “peepers”, which are polycarbonate dialysis membrane

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