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High metal reactivity and environmental risks at a site contaminated by glass waste

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HIGHLIGHTS

- Toxic metals are leached from glass waste when exposed in the environment.
- In soil/landfill samples, these metals dominate in geochemically active forms.
- The metal retention in the unsaturated zone is significant, at least in the short-term.
- Groundwater data and batch leaching tests, however, indicate on-going mobilization.
- Metal mobilization from glass waste should be considered associated with significant risks.

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ABSTRACT

This study addresses the reactivity and risks of metals (Ba, Cd, Co, Cr, Cu, Ni, Pb, Zn, As and Sb) at a Swedish site with large glass waste deposits. Old glassworks sites typically have high total metal concentrations, but as the metals are mainly bound within the glass waste and considered relatively inert, environmental investigations at these kinds of sites are limited. In this study, soil and landfill samples were subjected to a sequential chemical extraction procedure. Data from batch leaching tests and groundwater upstream and downstream of the waste deposits were also interpreted. The sequential extraction revealed that metals in <2 mm soil/waste samples were largely associated with geochemically active fractions, indicating that metals are released from pristine glass and subsequently largely retained in the surrounding soil and/or on secondary mineral coatings on fine glass particles. From the approximately 12 000 m^3 of coarse glass waste at the site, almost 4000 kg of Pb of Pb is estimated to have been lost through corrosion, which, however, corresponds to only a small portion of the total amount of Pb in the waste. Metal sorption within the waste deposits or in underlying soil layers is supported by fairly low metal concentrations in groundwater. However, elevated concentrations in downstream groundwater and in leachates of batch leaching tests were observed for several metals, indicating on-going leaching. Taken together, the high metal concentrations in geochemically active forms and the high amounts of as yet uncorroded metal-rich glass, indicate considerable risks to human health and the environment.

1. Introduction

Total metal concentrations are often used to assess the contamination level and potential risk at contaminated sites, and may indeed be useful for a first evaluation. However, it is well known that total concentrations provide limited information about the potential for metal leachability, bioavailability, and risk (Kördel et al., 2013; McLaughlin et al., 2000; Meers et al., 2007).

Landfills with glass waste constitute one example of sites where high total concentrations of heavy metals may be observed, but where the potential metal leachability and mobility has not been evaluated and discussed from a risk perspective. Today, as well as historically, glass constitutes an important waste fraction worldwide, quantitatively speaking. In the US, for example, 4.5% of the total 254 million t waste generated in 2013 was glass waste (US EPA, 2016). For the assessment of risks, historic landfills are, however, of

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greater concern than new ones; firstly, because the use of toxic substances in glass manufacturing was more poorly controlled in the past, and secondly, because waste generated prior to legislative controls on landfill management was deposited in landfills from which leaching to the surrounding environment may now occur. Although glass waste is generally considered to be inert, meaning that it can be assumed to "*not undergo any significant physical, chemical or biological changes*" and has a "*total leacheability … [that] … is insignificant*" (NFS, 2004:10; SFS, 2001:512), historical production of art glass and exclusive household glass was often associated with the use of toxic metals. The full leaded crystal glass produced in Sweden, for example, contained a minimum of 26 wt% lead oxide (Magnusson, 1971). Barium and Zn later replaced Pb. Arsenic trioxide, and later also Sb, was used as a decolorant and to prevent blistering, and a range of other metal oxides (e.g. of Cd, Co, Cr, Cu, and Ni) were used as colorants (Hermelin and Welander, 1986; Magnusson, 1971). So, depending on the character of the glass, glass waste may contain high concentrations of a variety of metals, but there are surprisingly few reports in the scientific literature on the leaching behavior of toxic metals from soil/glass waste matrices in natural environments. Those studies that have been conducted, however, indicate that metals may indeed be leached from glass in natural environments (Colomban et al., 2006; Doménech-Carbó et al., 2006; Silvestri et al., 2005; Sterpenich and Libourel, 2001).

The "Kingdom of Crystal" in southeastern Sweden is internationally recognized for its long tradition of crystal and art glass production. Glass has been produced in the area at more than 50 sites for over 300 years, resulting in large amounts of metals in soils and landfills. For example, initial consultant investigations of 22 glassworks sites in the region have estimated the total amounts of Pb, As and Cd to be approximately 3100, 420 and 30 tonnes, respectively, with remediation costs expected to exceed 100 million € (Höglund et al., 2007).

The aim of the present study was to assess the potential reactivity of metals at an old glassworks site in this area, and its implications in a short term (over a few decades) risk perspective. This was done by a characterization of the fractionation of a range of typical glass-production cation forming metals (Ba, Cd, Co, Cr, Cu, Ni, Pb and Zn) in soil samples from different subareas, with different glass waste loads, using a sequential extraction procedure. Secondary data from groundwater sampling wells, installed both upstream and downstream of the main waste deposits, and from batch leaching tests, were also retrieved and interpreted.

2. Materials and methods

2.1. Study site: Pukeberg as a representative for glassworks in the region

The study site chosen is Pukeberg in Nybro municipality, one of the most thoroughly investigated sites in The Kingdom of Crystal. Sites in this region contain large amounts of glass waste that is rich in heavy metals, and they are further of great interest from a risk perspective as they are generally located in villages where residents may grow their own vegetables and some extract drinking water from private wells. Many sites also border forested recreational areas of high accessibility, with edible berries and mushrooms. It is well known that many of the metals that were used in historical glass production have negative health effects (Järup and Åkesson, 2009; Silbergeld, 2003; Staessen et al., 1996; Åkesson et al., 2014), and studies of glass factory workers have assessed that occupational exposure during glass production increases the risk of cancer (IARC, 1993; Wingren, 1991). The extent to which local residents are affected is, however, not yet known. Understanding the mobility of contaminants at these sites thus has direct implications for the management of health risks.

Pukeberg is located on the northern part of the Nybro ridge, which is one of the largest glaciofluvial deposits and groundwater aquifers in southeastern Sweden. The groundwater level is typically found 1.5–3.0 m below ground surface. Although the glass waste is for the most part found above this level, high total metal concentrations have been found down to 2.5 m depth. The general ground water flow direction is towards the east and north-east. See also Fig. 1.

Glass production at Pukeberg started in 1871, and according to an inventory from 1970, approximately 450 t of glass waste was generated annually at that time and deposited in the landfill(s) on site (Elert and Höglund, 2012). From 1977 onwards, however, the waste has been deposited in external landfills, meaning that even the most recent waste currently present on the site has aged at least 35 years. The main landfill at Pukeberg covers an area of ca 15,000 m^2, with a maximum depth of ca 2.5 m, but smaller deposits of glass waste are scattered all over the site. To date 300 soil and landfill samples with a varying mix of glass waste have been collected and analysed, revealing severe contamination (Elert and Höglund, 2012).

2.2. Soil samples for sequential extractions

Soil with a varying mix of glass waste was collected from five subareas in October 2012 (Fig. 1). The subareas were selected based on differences in the content of visible glass waste (and the expected total metal concentrations) and in vegetational cover. In general, the glass waste is evenly mixed with the soil, although to different degrees and down to different maximum depths at the various subareas. Surface soil was in focus in the present study, due to its relevance from a risk perspective; for plant uptake, direct exposure via oral intake, or emission of dust particles (that can be inhaled). At each subarea, five pits were dug and from each pit a composite sample was taken, representing 0–40 cm, excluding the organic top layer when present, resulting in 25 samples in total. Below is a brief summary of the subareas:

Subareas A and B are landfills, with subarea B being the main landfill (Fig. 1). Different kinds of waste have been discarded here: glass cullets, batch residues, grinding sludge, ashes, oven bricks, crucibles, demolition remains etc. Both subareas are today covered by soil and dense vegetation that has accumulated since the landfills were closed. Samples from these two subareas consist of soil with a very high fraction of glass waste.

Subarea C is located beside the building where the glassworks furnaces were located and where the raw materials for the glass batches were stored and mixed. This subarea is covered by thin patches of grass. A small, but clearly visible, amount of glass waste sheets can be found when digging in the area.

Subarea D is located next to the main grindery and consists of filling material with a clear glass waste component. The area is covered by a grass lawn.

Subarea E is located at the northern end of the property. There is little visible glass at this spot, and the main source of contamination is grinding sludge, which has been deposited within the area and mixed with the natural soil.

2.2.1. Sample preparation and sequential extraction

The soil/glass waste samples were sieved to < 2 mm before analyses of metal concentrations, pH, water content and organic matter. The pH value was determined after shaking 25.0 g of fresh soil with 50 ml deionized water for 2 h (Reeuwijk, 2002). The soil water content was determined by drying fresh soil samples until constant weight at 105 °C, and the content of organic matter was