



Characterization and evaluation of arsenic and boron adsorption onto natural geologic materials, and their application in the disposal of excavated altered rock



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ABSTRACT

Construction of tunnels in Hokkaido, Japan often excavates rocks containing substantial amounts of arsenic (As) and boron (B). When these rocks are exposed to the environment, As and B are leached out that could potentially contaminate the surrounding soil and groundwater. Natural geologic materials contain minerals like Al-/Fe-oxyhydroxides/oxides that have As and B adsorption capabilities. Because these materials are widespread and readily available, they could be utilized in the mitigation of As and B leached out from these sources. This paper describes the ability of three natural geologic materials (i.e., pumiceous tuffs, partly-weathered volcanic ashes and coastal marine sediments) to sequester As and B from aqueous solutions and the actual leachate of a hydrothermally altered rock. The adsorption of As fitted well with either the Langmuir or Freundlich isotherm while that of B followed the Henry-type model (linear). Among the samples, those containing substantial amorphous Al and Fe exhibited higher As adsorption. However, the distribution coefficient of B only had a moderate positive correlation with these amorphous phases. The best adsorbent among these natural geologic materials was utilized in the adsorption layer of the column experiments. Adsorption of As was more effective the thicker the adsorption layer, but this retardation was only temporary due to significant changes in the pH. In contrast, the adsorption layer only retarded the migration of B to a limited extent.

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1. Introduction

Arsenic (As) and boron (B) are toxic at high concentrations and could cause a variety of human health and developmental problems. Chronic ingestion of trace amounts of As could cause arsenicosis, keratosis and cancers of the lungs, skin, bladder and kidneys (Chakraborty and Saha, 1987; Chen et al., 1992). On the other hand, B is an essential micronutrient, but has been reported to cause reproductive and developmental abnormalities at large doses (Fail et al., 1998). In nature, both of these elements are found only in trace amounts, but they are sometimes concentrated in certain geological features and anomalies. For instance, volcanic activities could cause hydrothermal alteration

and the subsequent enrichment of rocks with As and heavy metals (Pirajno, 2009).

Hydrothermally altered rocks, which are abundant in Japan, are formed underground so that they usually do not pose any environmental problems. Unfortunately, recent tunnel projects for roads and railways have excavated these rocks exposing them to the environment. If not disposed of properly, sulfide minerals in the rocks are oxidized and weathered resulting in the release of hazardous elements to the surrounding soil and groundwater. At the moment, they are disposed of in landfills with special liners similar to those used for municipal solid and industrial wastes (Katsumi et al., 2001; Lundgren and Soderblom, 1985; Wijeyesekera et al., 2001). However, this approach on the long term is not economically sustainable because of its prohibitively high cost in conjunction with the large volume of rocks excavated. In search of an alternative mitigation approach, we have studied in detail the leaching behavior and release mechanisms of several hazardous elements present in altered rocks (Tabelin and Igarashi, 2009). We have also found that altered rocks could partly mitigate the leaching of As through its adsorption onto precipitated iron (Fe)- and aluminum

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(Al)-oxides/oxyhydroxides (Tabelin et al., 2012b). However, the adsorption capabilities of these inherent minerals were insufficient to lower the concentration of As below the environmental standard of Japan (10 µg/L) (Tabelin et al., 2012b,c). Based on these previous results, a viable countermeasure to mitigate the leaching of As is to enhance the sequestration capability of altered rocks through the addition of suitable adsorbents.

Mohan and Pittman (2007) provided a comprehensive review of As adsorbents used in water and wastewater treatments. However, most of these studies pertain to either synthetic materials/minerals or naturally occurring materials that are composed of only a single mineral (e.g., natural hematite, bentonite and kaolinite). Thus, studies pertaining to the adsorption of As and B onto naturally occurring materials with complex mineral compositions are still lacking. Likewise, adsorption of As and B onto single component/mineral systems could not be used to predict the transport of these elements in multi-component systems like rocks and soils.

In this study, we evaluated As and B adsorption onto pumiceous tuff, partly-weathered volcanic ash and coastal sediments using classical batch adsorption experiments. Because most of these samples have trace amounts of As, the leachability of this geogenic As as a function of pH was also elucidated in selected samples. The best adsorbent was selected based on the leaching and adsorption results, and used to mitigate the leaching of As and B from an actual hydrothermally altered rock. This was done using column experiments with a crushed rock-bottom adsorption layer configuration (Tatsuhara et al., 2012), which is similar to the concept of permeable reactive barriers (PRB). Finally, the migration of As and B was simulated using the one-dimensional advection-dispersion with retardation equation to provide insights into the transport phenomena in the adsorption layer.

2. Materials and methods

2.1. Sample collection, preparation and characterization

The hydrothermally altered rock sample was collected from a road tunnel built in the central part of Hokkaido, Japan. The rock excavated in this area is mainly composed of partly altered mudstone and sandstone of marine origin formed during the Cretaceous period. The rock sample was collected from an interim storage site, which was built to accommodate freshly excavated rocks from the tunnel prior to their final disposal. Thus, the rock was partly oxidized due to its exposure to the environment for ca. 6 months. Sampling was done using shovels at random points around the interim storage site with collected samples varying in sizes from gravel (>20 mm in diameter) to silty sand (<2 mm in diameter). The rock sample was brought to the laboratory, air dried at room temperature, crushed using a jaw crusher and sieved through a 2 mm aperture screen. The <2 mm fraction was collected, mixed thoroughly and stored in air-tight containers to minimize its exposure to moisture. During tunnel construction, the rocks excavated usually have a wide distribution of sizes ranging from large boulders to very fine silt and clay. We chose to evaluate the <2 mm fraction because it represented the most reactive fraction of the bulk excavated rock. The chemical composition and mineralogical properties of this rock have been reported previously (Tabelin et al., 2012a,d). It is composed predominantly of silicate minerals (i.e., quartz and plagioclase), chlorite and calcite as minor minerals, and trace amounts of pyrite. It contains As and B at 6.9 and 113 mg/kg, respectively. In terms of its particle size distribution, the altered rock sample used in the columns is classified as loamy sand, which is composed of 89.4% sand, 5.3% fine sand, and 5.3% silt and clay.

Eleven natural geologic materials were collected for the experiments: three pumiceous tuffs (T-1, 2 and 3), two partly-weathered volcanic ashes (A-1 and A-2) and six coastal sediments (S-1–6). The three pumiceous tuffs and one volcanic ash (T-1–3; A-1) originated from the previous eruptions of Mt. Tokachi located around Obihiro

City (central part of Hokkaido). The other volcanic ash sample (A-2) came from the town of Kucchan (western part of Hokkaido) while all six coastal sediments were obtained near Hakodate City (southern part of Hokkaido). A brief description of these materials is summarized in Table 1. Samples of these geologic materials in their undisturbed state were collected using stainless steel cylinders for the determination of their hydraulic conductivities. Additional samples were obtained using hand shovels, air dried at room temperature, lightly crushed using mortar and pestle and sieved through a 2 mm aperture screen. The <2 mm fraction was utilized in the adsorption, leaching and column experiments. Chemical and mineralogical analyses were carried out on pressed powders of the samples (<50 µm) using an X-ray fluorescence spectrometer (Spectro Xepos, Rigaku Corporation, Japan) and an X-ray diffractometer (MultiFlex, Rigaku Corporation, Japan), respectively. Other important properties of these materials like particle size distribution and particle density were also measured. Their amorphous Al and Fe contents were determined by acidic oxalate solution extraction (McKeague and Day, 1965; Tamm, 1922), which was done by mixing 1 g of sample and 100 ml of acidic oxalate solution for 4 h at room temperature. The acidic oxalate solution was a 1:0.75 mixture of 0.23 M ammonium oxalate (C₂H₈N₂O₄) and 0.28 M oxalic acid (H₂C₂O₄). The zeta potential of the adsorbent used in the column experiments was measured using Nano-ZS60 (Malvern Instruments, UK). This analysis was done on the <50 µm fraction using 0.1 M hydrochloric acid (HCl) or sodium hydroxide (NaOH) solution for pH adjustment.

2.2. Batch experiments

2.2.1. pH dependent leaching experiments

Batch leaching experiments were conducted under ambient conditions by mixing 15 g of selected natural geologic samples (<2 mm) and 150 ml of prepared leachants. HCl and NaOH solutions of varying concentrations were used as leachants. The deionized water (18 M Ω · cm) used during the leachant preparation was obtained from a Millipore Milli-Rx 12α system (Merck Millipore, USA). After 24 h, the pH and redox potential (Eh) of the suspensions were measured followed by filtration of these suspensions through 0.45 µm Millex® sterile membrane filters (Merck Millipore, USA). All filtrates were acidified (pH < 2) and stored at 6° C prior to the chemical analyses.

2.2.2. Arsenic and boron adsorption experiments

Batch adsorption experiments were done by mixing solutions of known arsenate (As[V]) or B concentration with various amounts of the natural geologic samples at 120 rpm for 24 h. We only evaluated the adsorption of As[V] onto these natural materials because majority of As leached from the hydrothermally altered rock used in this study was As[V] (Igarashi et al., 2013). As[V] was prepared from reagent grade Na₂HAsO₄ · 7H₂O powder while B was prepared from 1000 mg/L standard solutions for atomic absorption spectrometry (Wako Pure Chemical Industries Ltd., Japan). The leachate samples were collected

Table 1
Description of the natural geologic materials and their sampling locations.

Sample	Description	Category
T-1	Pumiceous tuff	Loamy sand
T-2	Pumiceous tuff	Loamy sand
T-3	Pumiceous tuff	Loamy sand
A-1	Partly-weathered volcanic ash	Loamy sand
A-2	Partly-weathered volcanic ash	Clay loam
S-1	Coastal marine sediment	Sandy loam
S-2	Coastal marine sediment	Loamy sand
S-3	Coastal marine sediment	Conglomeratic sandy loam
S-4	Coastal marine sediment	Conglomeratic sandy loam
S-5	Coastal marine sediment	Sandy loam
S-6	Coastal marine sediment	Loamy sand

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