



Short and long term release mechanisms of arsenic, selenium and boron from a tunnel-excavated sedimentary rock under in situ conditions



Shuichi Tamoto^{a,b}, Carlito Baltazar Tabelin^{b,*}, Toshifumi Igarashi^b, Mayumi Ito^b, Naoki Hiroyoshi^b

^a Civil Engineering Research Institute for Cold Region, Public Works Research Institute, Sapporo, Japan

^b Division of Sustainable Resources Engineering, Faculty of Engineering, Hokkaido University, Sapporo, Japan

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ABSTRACT

Sedimentary rocks of marine origin excavated from tunnel construction projects usually contain background levels of hazardous trace elements, but when exposed to the environment, they generate leachates with concentrations of arsenic (As), selenium (Se) and boron (B) exceeding the WHO guideline for drinking water. In this study, the leaching of As, Se and B was evaluated under in situ conditions at various flow patterns, particle size distributions and column thicknesses. The results showed that these trace elements were leached out of the rock via short and long term mechanisms. In the short term, all three elements were rapidly and simultaneously released due to the dissolution of soluble evaporite salts formed from entrapped sea water of the Cretaceous. After their rapid release, however, these trace elements behaved differently as a result of their contrasting adsorption affinities onto minerals like clays and Fe-oxyhydroxides, which were further influenced by the pH, presence of coexisting ions and speciation of the trace elements. Selenium was quickly and easily transported out of the columns because it was mostly present as the very mobile selenate ion (Se[VI]). In comparison, the migration of As and B was hindered by adsorption reactions onto mineral phases of the rock. Boron was initially the least mobile among the three because of its preferential adsorption onto clay minerals that was further enhanced by the slightly alkaline pH and high concentrations of Ca²⁺ and Na⁺. However, it was gradually re-mobilized in the latter part of the experiments because it was only weakly adsorbed via outer sphere complexation reactions. In the long term, the rock continued to release substantial amounts of As, Se and B via pyrite oxidation and adsorption/desorption reactions, which were regulated by the temperature and rainfall intensity/frequency on site.

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1. Introduction

Arsenic (As), selenium (Se) and boron (B) are among the most common and persistent trace inorganic contaminants in the environment that are strictly regulated because of their

dose-dependent toxicities. Arsenic is well-known for its lethal and carcinogenic properties (Cebrian et al., 1983; Chakraborty and Saha, 1987; Chen et al., 1985, 1992; Smith et al., 1992) while Se and B, though essential micronutrients, are toxic at high concentrations. Selenium is a key component of glutathione peroxidases, enzymes vital in the metabolism and detoxification of oxygen, while B is essential during calcium metabolism and utilization (Levander et al., 1983; Navarro-Alarcon and Cabrera-Vique, 2008; Nève, 1991; Rotruck et al., 1973). In controlled dosages, Se and B also have important

* Corresponding author. Tel./fax: +81 11 706 6315.

E-mail addresses: 95353@ceri.go.jp (S. Tamoto), carlito@eng.hokudai.ac.jp (C.B. Tabelin), tosifumi@eng.hokudai.ac.jp (T. Igarashi), itomayu@eng.hokudai.ac.jp (M. Ito), hiroyosi@eng.hokudai.ac.jp (N. Hiroyoshi).

medicinal uses. Selenium is used to reduce heavy metals and xenobiotic toxicity (Ganther, 1980), prevent the endemic fatal cardiomyopathy called “Keshan” disease (Yang et al., 1983), and treat muscular dystrophy appearing in patients on long-term parenteral nutrition (Van Rij et al., 1979). Likewise, B prevents and treats various forms of arthritis, as well as improves psychomotor response, brain function and estrogen ingestion response in postmenopausal women (Havercroft and Ward, 1991; Nielsen, 1994; Nielsen et al., 1987, 1990; Shah and Vohora, 1990; Travers et al., 1990). In excess, however, Se increases the risks of developing breast, colorectal and kidney cancers, melanoma and lymphoid neoplasms, Parkinson's disease, and amyotrophic lateral sclerosis (ALS) (Ayaz et al., 2008; Brtko and Filipcik, 1994; Chatterjee and Banerjee, 1982; Maraldi et al., 2011; Stoica et al., 2000; Vinceti et al., 1995, 1996, 1998, 2013) while B causes nausea, vomiting, diarrhea, abdominal cramps, erythematous lesions on the skin and mucous membranes, circulatory collapse, tachycardia, cyanosis, delirium, convulsions, and coma (Beyer et al., 1983; Siegel and Wason, 1986).

Hazardous trace elements are sometimes naturally concentrated in rocks, soils and sediments as a result of certain geological processes and anomalies. One such process is the hydrothermal alteration of rocks, which occurs when rocks and solute-rich superheated solutions/fluids interact with each other. This process transforms ordinary rocks into hydrothermally altered rocks enriched with As and heavy metals (Pirajno, 2009; Tabelin et al., 2012b). Hydrothermal alteration of rocks is a widely studied topic because of its importance in the formation of precious and base metallic ores. On an environmental perspective, however, such rocks are hazardous and potential sources of toxic inorganic contaminants. In our previous studies, we conducted batch, column and in situ experiments to understand the leaching behavior and release mechanisms of several trace elements (e.g., As and lead (Pb)) contained in such rocks as well as proposed ways of mitigating their negative impacts to the environment (Tabelin and Igarashi, 2009; Tabelin et al., 2010; Tabelin et al., 2012a, 2012b, 2012c; Tatsuhara et al., 2012; Tabelin et al., 2013; Tabelin et al., 2014a).

More recently, we have identified marine sedimentary rocks as another naturally occurring material that are potential sources of hazardous trace contaminants. Although marine sedimentary rocks generally have low concentrations of trace elements (i.e., close to background levels), they are capable of generating leachates with concentrations of hazardous trace elements (e.g., As, Se and B) exceeding the environmental standards of Japan and the WHO guideline for drinking water (Tabelin et al., 2014b, 2014c). These results were quite disturbing given the facts that sedimentary rocks are widespread in nature and make up a significant portion of most aquifer systems. Numerous studies have been conducted on sedimentary rocks especially those pertaining to their diagenesis, stratigraphy, mineralogical composition and physical properties (Bjørlykke, 2013; Prezbindowski and Tapp, 1991), but hardly any have been done to understand how hazardous trace elements behave when these rocks are exposed to the environment.

In our previous studies, we reported that the leaching behaviors of As, Se and B in marine sedimentary rocks largely depended on the stability of minerals/phases containing the

bulk of these elements (i.e., solid-phase partitioning) and the pH of the rock–water system (Tabelin et al., 2014b, 2014c). The mobilities of As and Se, for instance, were regulated by processes like dissolution of soluble phases, adsorption/desorption and pyrite oxidation, the extents of which varied depending on the pH of the rock–water system (Tabelin et al., 2014c). These previous studies, though insightful, were done using batch reactor-type experiments whose results do not provide much about how trace elements behave with variations in temperature, rainfall, infiltration rate and water content. Such variations are prevalent in the actual field setting and essential for the appropriate management of these rocks.

To address these issues, we conducted column experiments under in situ conditions for 821 days. In these experiments, we evaluated how water saturation/content, particle size distribution and rock bed thickness affected the leaching of As, Se and B from a sedimentary rock sample of marine origin. Throughout the in situ experiments, we continuously monitored changes in temperature, volumetric water content (θ), partial pressure of oxygen (PO_2) and electrical conductivity (EC) within the columns as well as the evolutions of As, Se, B and coexisting ions in the effluents. Furthermore, geochemical modeling was used to predict the speciation of trace elements and provide insights into the stability of mineral phases/constituents of the rock (e.g., Fe-oxyhydroxides and clay minerals).

2. Materials and methods

2.1. Sedimentary rock of marine origin

The sedimentary rock sample used in this study was a mixture of mudstone and sandstone formed during the Cretaceous. It was collected from the interim storage area of a road-tunnel project in northern Hokkaido, Japan. The chemical and mineralogical properties including the solid-phase partitioning of As, Se and B in this rock were reported in our previous studies (Tabelin et al., 2014b, 2014c). Quartz and plagioclase are the main mineral components of this rock (Supplementary Fig. 1). Calcite, chlorite and mica were also detected in minor quantities while pyrite was found in trace amounts. This rock contains 9.059 and 113 mg/kg of As, Se and B, respectively (Tabelin et al., 2014b), which are all within the reported average concentration ranges of these elements in sedimentary rocks and soils (Fishbein, 1983; Girling, 1984; Ishikawa and Nakamura, 1993; Spears, 1964; Webster, 1999). The majority of As and Se in this rock was partitioned with the exchangeable (As: 3.1 mg/kg; Se: 0.24 mg/kg) and sulfide phases (As: 3.4 mg/kg; Se: 0.15 mg/kg) (Tabelin et al., 2014c). In comparison, B was mostly associated with the refractory crystalline/residual phase (75 mg/kg). However, amounts of this element partitioned with the exchangeable (18 mg/kg) and carbonate (9.3 mg/kg) phases were still significant.

2.2. Column experiments

2.2.1. Apparatus

The in situ column setup used in this study is illustrated in Fig. 1. The cylindrical tubes are made of PVC with an inner diameter of 298 mm and a height of 780 mm. They were mounted on an insulated-steel box bolted on a solid concrete slab. After construction of the columns, fine-meshed PVC

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