



Potential for leaching of arsenic from excavated rock after different drying treatments



Jining Li ^a, Tomoya Kosugi ^a, Shohei Riya ^a, Yohey Hashimoto ^b, Hong Hou ^c, Akihiko Terada ^a, Masaaki Hosomi ^{a,*}

^a Department of Chemical Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Naka, Koganei, Tokyo 184-8588, Japan

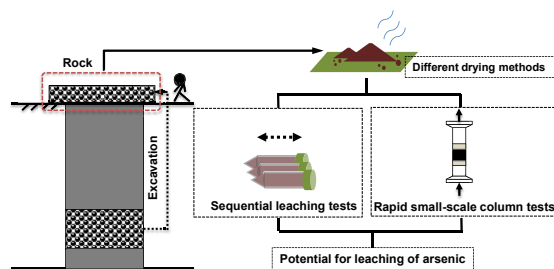
^b Department of Bioapplications and Systems Engineering (BASE), Tokyo University of Agriculture and Technology, 2-24-16 Naka, Koganei, Tokyo 184-8588, Japan

^c State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Dayangfang 8, Beijing 100012, PR China

HIGHLIGHTS

- Considerable total As was water-leached from an excavated rock.
- Air-, 40 °C-, and freeze-drying treatments increased the As leaching.
- Freeze-drying increased As leaching by the largest extent.
- Dissolution of Fe, Al, and dissolved organic carbon correlated with As leaching.
- Pollution potential indices are proposed to evaluate the pollution threat of As.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 21 January 2016

Received in revised form

19 March 2016

Accepted 28 March 2016

Handling Editor: X. Cao

Keywords:

Arsenic
Excavated rock
Leaching
Drying

ABSTRACT

Leaching of arsenic (As) from excavated rock subjected to different drying methods is compared using sequential leaching tests and rapid small-scale column tests combined with a sequential extraction procedure. Although the total As content in the rock was low (8.81 mg kg^{-1}), its resulting concentration in the leachate when leached at a liquid-to-solid ratio of 10 L kg^{-1} exceeded the environmental standard ($10 \mu\text{g L}^{-1}$). As existed mainly in dissolved forms in the leachates. All of the drying procedures applied in this study increased the leaching of As, with freeze-drying leading to the largest increase. Water extraction of As using the two tests showed different leaching behaviors as a function of the liquid-to-solid ratio, and achieved average extractions of up to 35.7% and 25.8% total As, respectively. Dissolution of As from the mineral surfaces and subsequent re-adsorption controlled the short-term release of As; dissolution of Fe, Al, and dissolved organic carbon played important roles in long-term As leaching. Results of the sequential extraction procedure showed that use of $0.05 \text{ M } (\text{NH}_4)_2\text{SO}_4$ underestimates the readily soluble As. Long-term water extraction removed almost all of the non-specifically sorbed As and most of the specifically sorbed As. The concept of pollution potential indices, which are easily determined by the sequential leaching test, is proposed in this study and is considered for possible use in assessing efficacy of treatment of excavated rocks.

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* Corresponding author.

E-mail address: hosomimasaaki1130@gmail.com (M. Hosomi).

1. Introduction

Arsenic (As), a metalloid belonging to Group 15 of the Periodic Table, is ubiquitously present in the natural environment at trace levels (Wilson et al., 2010). According to a report of the World Health Organization (WHO), As is one of ten chemicals of major public health concern (WHO, 2012). Long-term As exposure from food and drinking water can cause cancer and skin lesions, as well as developmental effects, cardiovascular disease, neurotoxicity, and diabetes (WHO, 2012). Because of these negative impacts on human health, the WHO recommended a provisional standard of $10 \mu\text{g L}^{-1}$ for arsenic in drinking water (WHO, 1994).

Many construction projects require excavation of large quantities of deep rocks. Although the amounts of As in these rocks are often very low, the concentrations in their resultant leach solutions can exceed environmental standards: for example, Tabelin et al. (2014a) found that the As concentrations in leachates from some excavated rocks were higher than the Japanese environmental standard ($10 \mu\text{g L}^{-1}$). This underscores the need to properly dispose of such rocks. In fact, in Japan, efforts are now directed at the potential risks of excavated rocks, and the Soil Contamination Countermeasures Law of Japan (Ministry of the Environment, 2011) was amended to extend the interpretation to cover these solids. To assess the potential for contamination of As from such rocks and to develop effective and economically feasible remediation technologies, more detailed knowledge about As behavior in excavated rocks is required to better understand its release to the surrounding environment.

To date, the risk of pollutants leaching from a variety of solids has been determined by laboratory leaching tests, such as batch and column tests (Grathwohl and Susset, 2009). Batch tests provide a snapshot of a particular liquid-to-solid (L/S) ratio (Maszkowska et al., 2013), but do not simulate actual field conditions because of friction between the particles during mechanical agitation, centrifugation, and filtration of the sample prior to analysis. Column test conditions are more similar to field conditions and more representative than batch tests because there is no mechanical friction, the packed material has self-filtration capacity, and observation of time-dependent leaching behavior is possible (Krüger et al., 2012). Although some studies have compared batch and column tests with respect to As release from soils or wastes (Al-Abed et al., 2008; Kim and Hyun, 2015), no study has quantitatively compared these techniques for investigating the potential for As release from excavated rocks.

An appropriate methodology for storage of excavated rocks still requires development. As it acts on a soil sample, drying treatment plausibly enables a rock sample to be stored for longer time and contributes to development of a standardized methodology, which allows one to compare results for As release from different rocks. However, drying may change the status and properties of trace elements in the rocks and raises the question of whether As release from dried samples is representative of field conditions (Supriatin et al., 2015). To date, the effect of different drying methods on As extraction from soils has been studied (Huang et al., 2015), however, the influence of drying on As leaching behavior from excavated rocks is largely unknown. The objectives of this study are, therefore: (1) to investigate the effects of drying methods on the As leaching from an excavated rock; (2) to compare the potential for As leaching under batch and column conditions; and (3) to identify the dominant processes governing As leaching from this excavated rock.

2. Materials and methods

2.1. Sample collection and characterization

The excavated rock used in our study was obtained from Tokyo,

Japan, in February 2015. After collection, all samples were placed in airtight polyethylene bags and transported to the laboratory. The collected rock was pretreated by four common drying methods: (1) air-drying at room temperature for 7 days (AD); (2) oven-drying at 40°C for 24 h (40D); (3) freeze-drying at -50°C for 24 h (FD); and (4) no drying (ND). After being subjected to the selected pretreatment, the samples were gently crushed and sieved through a 2-mm sieve. A portion of the sample with a particle size of less than 2 mm was used for the leaching experiments. Further description of sample characterization is provided in the Supplementary Material. Selected physicochemical properties are shown in Table S1. The total As concentration in this rock was 8.81 mg kg^{-1} .

2.2. Leaching experiments

2.2.1. Sequential leaching tests

Sequential leaching tests (SLT) are sometimes used to study long-term leaching of contaminants from solids into water (Grathwohl and Susset, 2009). In this study, referring to Notification No. 46 of the Ministry of Environment (Japan), the SLT was conducted under ambient conditions. A 3-g sample (dry weight basis; 4.44 g for sample with ND) and 30 mL of deionized water ($18 \text{ M}\Omega \text{ cm}$) were mixed in a 50-mL centrifuge tube. To determine an appropriate shaking time, we performed preliminary batch leaching experiments with different shaking periods (0.5 h, 1 h, 3 h, 6 h, 12 h, and 24 h). The mixture was shaken at a rate of 200 rpm at room temperature and pressure. The slurry was centrifuged at about 4700 g for 15 min and the supernatant was filtered through a $0.45\text{-}\mu\text{m}$ membrane filter before analysis. These procedures were repeated until the As concentration in the leachate was below its detection limit ($0.05 \mu\text{g L}^{-1}$). The residual sample material was dried using the methods described above and then digested for As measurement.

After the $0.45\text{-}\mu\text{m}$ filtration, As in leachates is commonly classified into dissolved and colloidal fractions (Nystrand et al., 2012). In our study, part of the $0.45\text{-}\mu\text{m}$ filtered leachate after 24-h shaking was further filtered using a $0.1\text{-}\mu\text{m}$ filter and the As in this filtrate was considered to be present in dissolved form. The difference between the $0.45\text{-}\mu\text{m}$ and $0.1\text{-}\mu\text{m}$ filterable As was attributed to colloidal form. Fe and Al in the leachates were also classified by the same method.

2.2.2. Rapid small-scale column tests

Rapid small-scale column tests (RSSCT), which allow breakthrough curves as functions of time and volume of permeates to be generated, were originally developed to simulate the removal of organic pollutants and natural organic matter by granular activated carbon (Crittenden et al., 1987, 1991). They are appropriate for use with the excavated rock in this study because of its high permeability. A series of stand-alone RSSCT columns (internal diameter, 1.7 cm; length, 24 cm) were constructed and an up-flow configuration was employed, which differs from the regular RSSCT test. The experimental set-up is shown in the Supplementary Material. To readily compare these As leaching results with those of the SLT, a 3.0-g sample (4.44 g for sample with ND) was packed in each column. The column was first slowly saturated with water from bottom to top; water was then continuously supplied at a flow rate of 1.0 mL min^{-1} . Fifteen column effluent samples of 30 mL each (equivalent to 10 L kg^{-1} of L/S) were collected (sample numbers 1–15); thereafter, samples were collected every 50 mL (equivalent to every 16.7 L kg^{-1}) and numbered from 16 onward. Sampling continued until the As concentration was below its detection limit. The residual material was dried and digested for As measurement.

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