



# Arsenic-containing soil from geogenic source in Hong Kong: Leaching characteristics and stabilization/solidification



Jiang-Shan Li<sup>a</sup>, Jingzi Beiyuan<sup>a</sup>, Daniel C.W. Tsang<sup>a, \*</sup>, Lei Wang<sup>a</sup>, Chi Sun Poon<sup>a, \*\*</sup>,  
Xiang-Dong Li<sup>a</sup>, Scott Fendorf<sup>b</sup>

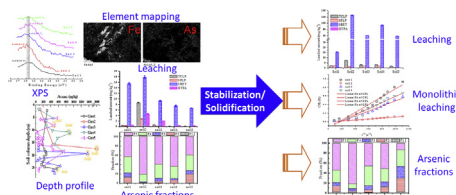
<sup>a</sup> Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China

<sup>b</sup> Earth System Science Department, Stanford University, Stanford, CA 94305, United States

## HIGHLIGHTS

- Elevated levels of As are encountered in deeper layer below ground surface.
- Despite high concentrations, geogenic As shows minimal leaching potential.
- The majority of geogenic As exist as As(V) strongly bound to Fe oxides.
- Stabilization/solidification provides physical encapsulation and prevents exposure.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Geogenic sources of arsenic (As) have aroused extensive environmental concerns in many countries. This study evaluated the vertical profiles, leaching characteristics, and surface characteristics of As-containing soils in Hong Kong. The results indicated that elevated levels of As ( $486\text{--}1985\text{ mg kg}^{-1}$ ) were mostly encountered in deeper layer (15–20 m below ground). Despite high concentrations, geogenic As displayed a high degree of chemical stability in the natural geochemical conditions, and there was minimal leaching of As in various leaching tests representing leachability, mobility, phytoavailability, and bioaccessibility. Microscopic/spectroscopic investigations suggested that As in the soils was predominantly present as As(V) in a coordination environment with Fe oxides. Sequential extraction indicated that the majority of As were strongly bound with crystalline Fe/Al oxides and residual phase. Yet, uncertainties may remain with potential As exposure through accidental ingestion and abiotic/biotic transformation due to changes in geochemical conditions. Hence, the effectiveness of stabilization/solidification (S/S) treatment was evaluated. Although the leached concentrations of As from the S/S treated soils increased to varying extent in different batch leaching tests due to the increase in alkalinity, the mobility of As was considered very low based on semi-dynamic leaching test. This suggested that As immobilization in the S/S treated soils was predominantly dependent on physical encapsulation by interlocking framework of hydration products, which could also prevent potential exposure and allow controlled utilization of S/S treated soils as monolithic materials. These results illustrate the importance of holistic assessment and treatment/management of As-containing soils for enabling flexible future land use.

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## 1. Introduction

Arsenic (As) contamination is one of the major environmental problems around the world, causing a global epidemic of poisoning,

\* Corresponding author.

\*\* Corresponding author.

E-mail addresses: [dan.tsang@polyu.edu.hk](mailto:dan.tsang@polyu.edu.hk) (D.C.W. Tsang), [chi-sun.poon@polyu.edu.hk](mailto:chi-sun.poon@polyu.edu.hk) (C.S. Poon).

with tens of thousands of people having developed skin lesions, cancers, and other symptoms due to natural sources and human activity-induced emissions (Fendorf et al., 2010; Chakraborti et al., 2016). Most of the reported studies in the remediation literature focused on soils contaminated by anthropogenic sources of As (Tsang et al., 2014; Cai et al., 2016; Fang et al., 2016; Ahmad et al., 2017), while the occurrence of geogenic As arouses major concern for groundwater quality in many countries (Fendorf et al., 2010; Stuckey et al., 2015). Natural sources of As include volcano release and weathering of arsenic-containing minerals and ores, which may interfere urban land development (Bondu et al., 2016). The commonly encountered range of As in soils lies between 0.2 and 40 mg kg<sup>-1</sup>, which depends on the soil horizon, type of soil, and lithology (Moreno-Jiménez et al., 2012).

The solid-state species of As is widely recognized to influence the bioaccessibility and fate of As in the environment, which is crucial for assessing potential risk and developing suitable remediation strategies (Tufano et al., 2008; Kim et al., 2014). Arsenite and arsenate species interchange oxidation states depending on redox potential, pH, geochemical composition, as well as abiotic and biotic transformation (Smedley and Kinniburgh, 2002; Bei yuan et al., 2017a). The major As species in the environment are predominantly oxyanions, of which As(III) is more mobile and toxic than As(V) as the latter can be strongly adsorbed onto the surfaces of Fe, Al, and Mn oxides in soils (Manning et al., 2002; Masue-Slowey et al., 2011; Ying et al., 2012; Tsang and Hartley, 2014; Zhang et al., 2015). As the total As concentration in the soil is not a good indicator of its potential environmental impact, previous studies have employed sequential extraction procedures and spectroscopic analysis for investigating the As speciation (Wenzel et al., 2001; Clancy et al., 2015) as well as different leaching methods for understanding the leachability, mobility, bioaccessibility, and phytoavailability of As in soils (Karak et al., 2011; Tsang et al., 2013a; Bei yuan et al., 2016; Chen et al., 2017).

To reduce environmental risks and recover land resource for flexible use, extensive efforts have been made for devising an appropriate remediation of As-containing soil. While chemical-enhanced soil washing resulted in undesirable increase in lability of residual As (Tsang et al., 2013b; Bei yuan et al., 2017b) and phytoremediation with hyperaccumulators was time-consuming and marginally effective (Mahar et al., 2016), stabilization/solidification (S/S) has been one of the most effective remediation methods for anthropogenic contamination of As (Leist et al., 2003; Dermatas et al., 2004). The binders react during hydration with arsenic to form stable calcium-arsenate precipitates, sorption onto calcium-silicate-hydrates, or substitution mechanisms within crystalline lattice of ettringite (Vandecasteele et al., 2002; Halim et al., 2004; Phenrat et al., 2007). However, the feasibility of S/S for immobilizing highly concentrated As-containing soils due to geogenic source requires additional investigation, especially for the change in fractionation and fate of residual As based on different leaching and spectroscopic tests.

Therefore, this study evaluated the depth profiles, fractionation, and leaching characteristics of geogenic As-containing soils in Hong Kong. The As speciation and surface characteristics was analyzed by spectroscopic and microscopic tests, while the operationally defined leachability, mobility, bioaccessibility, phytoavailability, and semi-dynamic leaching characteristics of As in soils were assessed before and after S/S treatment.

## 2. Materials and methods

### 2.1. Geogenic As-containing soils

Fifty four soil samples at 0.5–28 m below ground surface (every

0.5 m) from five selected boreholes were collected from Northeast New Territories of Hong Kong, China. The soil columns were sealed to be air-tight at the site, frozen with ice packs in freezing boxes during transportation, and stored at –20 °C in the dark until freeze-drying for subsequent microscopic and spectroscopic analysis. For S/S experiments, the soils were oven-dried at 60 °C and passed through a 2 mm sieve. The concentrations of total metals were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES, SPECTROBLUE) after strong acid digestion (1:4 concentrated HNO<sub>3</sub> and HClO<sub>4</sub> (v/v)) according to ASTM D3974-81. We included 20% randomly selected replicates, spiked samples, and reagent blanks for quality control of sample analysis, and used NIST Reference Soil 2710A (containing 1540 mg/kg As) for quality assurance. The recovery rate of As was 91–93% after digestion and ICP-OES analysis, indicating good recovery and reliability/reproducibility. Five soil samples at varying depths from different boreholes that contained high As concentrations (486–1985 mg kg<sup>-1</sup>) were selected for subsequent analysis. Based on X-ray diffractometer analysis (XRD, D8 Advance, Bruker), the main minerals of the soils were quartz, kaolinite, muscovite, and iron oxides. The organic matter content was measured by loss on ignition at 550 °C; the cation exchange capacity (CEC) was measured after percolation of 1.0 mol L<sup>-1</sup> ammonium acetate solution at pH 7. The grain size distribution was analyzed by wet sieving and hydrometer tests. The physico-chemical properties of the five soils are shown in Table 1.

Scanning electron microscopy (SEM) with elemental mapping (VEGA3, TESCAN) was conducted on freeze-dried, gold-coated soil samples to investigate the morphology and elemental distribution (As, O, and Fe) on the surface of As-containing soils. The speciation of geogenic As was characterized using Kratos Axis Ultra Imaging X-ray photoelectron spectroscopy (XPS) with monochromatic Al K $\alpha$  X-ray. A broadscan was obtained using 50 eV pass energy, while narrow high resolution scans of As 3d were obtained using 20 eV pass energy. The charge effect was corrected using the C 1s line at 285 eV. The spectra were fitted using curve-fitting program XPSPEAK 4.1 and a least-squares procedure with peaks of 20% of the Lorentzian-Gaussian peak shape after subtraction of Shirley baseline. The component peaks were identified by comparison of their binding energies with values in the literature (NIST, 2012).

### 2.2. Stabilization/solidification

The binder used in this study was ordinary Portland cement (OPC, ASTM Type I, 21% SiO<sub>2</sub>, 5.9% Al<sub>2</sub>O<sub>3</sub>, 64.7% CaO). To keep the same workability of the mixture, the S/S samples were prepared with a constant water-to-soil ratio of 0.3 for soils 2 and 3, a ratio of 0.4 for soil 1, and a ratio of 0.5 for soils 4 and 5, because of their variation in clay content and water absorption. The cement content was only 5 wt% for low alkalinity and economic considerations. The soil samples and binder were mixed uniformly for 1 min using a mechanical mixer before adding water. The blended paste was further mixed for another 2 min and cast into steel moulds (2 × 2 × 2 cm) with vibration. The samples were cured at 23 °C and 75% relative humidity for 24 h, and then the hardened S/S materials were removed from the moulds and cured in a steam cabinet at 60 ± 2 °C for 7 d. Unconfined compressive strength of the S/S product was measured by using a universal testing machine (Testometric CXM 500-50KN) at a loading rate of 0.3 mm min<sup>-1</sup> according to BS EN12390-3. The strength was compared with the criteria for reuse of S/S treated soils as fill materials for site formation in Hong Kong (i.e., ground levelling to the design formation level prior to construction works) (HK EPD, 2011). All the tests were duplicated (or triplicated where necessary) and the average results were reported.

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