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## Arsenic migration and transformation in aquifer sediments under successive redox oscillations

Ping Ni<sup>a,b</sup>, Huaming Guo<sup>a,b,1</sup>, Rongxiao Yuan<sup>a,b</sup>, Qin Huang<sup>b</sup>

<sup>a</sup>State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Beijing 100083, P.R. China

<sup>b</sup>School of Water Resources and Environment, China University of Geosciences (Beijing), P.R. China

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### Abstract

Sediments were taken from the Hetao basin to investigate effects of different lithologies and grain size distribution on As behaviors during oscillation of redox conditions. Total digestion and sequential extraction procedures showed that Fe and Mn oxide minerals played important roles for As accumulation. The co-variation of As, CIA (Chemical Index of Alteration), ICV (Index of Compositional Variability) and  $Al_2O_3/SiO_2$  indicated that As were enriched in strongly chemical weathering sediments weathered from mature mudrocks. Two sediments samples with the same As contents (21.9  $\mu\text{g/g}$ ) but different lithologies (fine sand versus clay) were chosen for redox oscillation experiments. As released from clayey (C-1) suspension was lower than those in C-2 (grounded sand) and C-3 (sand). Under reducing conditions, the maximum As concentration in C-1 was 9.99  $\mu\text{g/L}$ , however, it was 149  $\mu\text{g/L}$  in C-2 and 120  $\mu\text{g/L}$  in C-3. It inferred that the lithology and mineralogy play an important role in As fixation and mobilization. The grounded sands released more As in reducing conditions and sequestered more As in oxic conditions relative to the pristine sands.

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### 1. Introduction

It has been well-accepted that groundwater As originates from sediments due to reductive dissolution of Fe/Mn oxides<sup>1</sup>. Weathering and biological transformation of As containing minerals under natural conditions can mobilize As from rocks and sediments to the aquatic environment<sup>2,3</sup>.

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\* Corresponding author. Tel.: +86-10-82321366; fax: +86-10-82321081.  
E-mail address: [hmguo@cugb.edu.cn](mailto:hmguo@cugb.edu.cn)

Minerals, such as clay minerals and Fe(III) oxide minerals, are the most common constituents of the subsurface environment<sup>3</sup>. Many studies found that As is enriched in argillaceous deposits, due to the large specific surface of fine-grained particles<sup>4</sup> and high affinity of As to the large proportion of sulfide minerals, oxides, organic matter and clays<sup>5</sup>. It has been suggested that successive redox oscillations increase the crystallinity in Fe(III)-oxyhydroxides<sup>6</sup>, modify the structure of clays<sup>7</sup> and result in the passivation of mineral surfaces<sup>8</sup>. However, it is unclear how grain size distributions and mineralogical compositions affect As behaviors during successive redox oscillations. The objectives of this study are to: (1) figure out the relationship between sediment As and different As fractions; (2) characterize the migration and transformation of As, Fe, Mn and other trace elements under successive redox conditions; (3) assess effects of grain size distributions and mineralogical compositions on As mobility.

## 2. Method and materials

### 2.1. Field site characterization and sediments sampling

The study area is located in the Shaihai town in the western part of Hetao basin. In 2012, two boreholes were drilled to take sediment samples. One is located near a natural lake (BH02), and the other (BH01) in the up-gradient of BH02. One hundred and six sediment samples were taken from two boreholes at depths up to 90 m below land surface (bls). Sequential extraction procedure was carried out for sediment samples to characterize As forms in sediments, including exchangeable As (S1), strongly adsorbed As (S2), very amorphous Fe oxyhydroxides-bound As (S3), amorphous Fe oxyhydroxides-bound As (S4), crystalline Fe oxyhydroxides-bound As (S5), silicates-bound As (S6), pyrite-bound As (S7), and residual As (S8). The detailed procedure can be found in<sup>9</sup>.

### 2.2. Experimental setup

The experimental setup is comprised of three 2.5 L glass vessels with a heating jacket wrapped outside (Fig. 1). The heating jackets allow for temperature control. ORP and pH variations were continuously monitored by ORP and pH electrodes. Data were recorded via a data logger. The ORP was controlled by purging Ar (and/or injection of sodium ascorbate) or O<sub>2</sub>. C-1 is the first vessel with clayey (BH02-64) sediment (major grain size: 3-11 μm) inside, C-2 is the second vessel with grounded sand (BH01-11) sediment (major grain size: 3-35 μm) inside and C-3 is the third vessel with sand (BH01-11) sediment (major grain size: 100-300 μm) inside. These two sediment samples were chosen because they had same total As contents but different lithologies. Sediment samples were mixed with deionized water with a ratio of 250 g to 2 L in glass vessels. Argon with a small amount of sodium ascorbate (i.e., 0.12 mL 0.5 M sodium ascorbate) or O<sub>2</sub> were injected in the system to lower or increase redox potentials, respectively. The experiments were performed three redox oscillations, with each oscillation including 7 days of reducing conditions (ORP <-150 mV) and 5 days of oxidizing states (ORP >150 mV). During the test, ORP was controlled with the pH left free to evolve naturally to mimic conditions occurring in the field. The slurry was agitated continuously at 150 rpm. The temperature was controlled at 31°C.

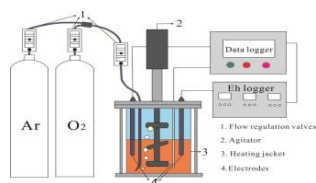


Fig. 1. The experimental setup for redox oscillation

## 3. Results and discussion

### 3.1. Impact of sedimentary provenance and weathering on arsenic distribution

Chemical Index of Alteration ( $CIA = (Al_2O_3 / (Al_2O_3 + CaO^* + Na_2O + K_2O)) \times 100$ ) is a chemical index, widely used to determine the degree of chemical weathering. As shown in Fig. 2, the CIA values were mostly between 50 and 60, which reflect that the source region had gone through weakly chemical weathering in cold and arid

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