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Interactions between arsenic adsorption/desorption and indigenous bacterial activity in shallow high arsenic aquifer sediments from the Jianghan Plain, Central China

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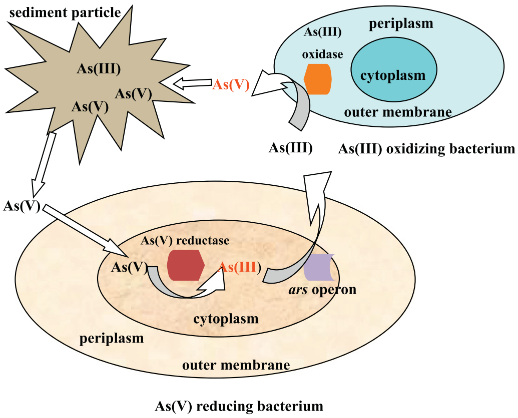
HIGHLIGHTS

- Jianghan Plain is a recently discovered high arsenic area in China.
- Bacteria affect arsenic adsorption/desorption and oxidation/reduction.
- Bacteria establish an arsenic dynamic equilibrium between sediment and groundwater.
- Arsenic adsorption/desorption is related to bacterial activity and sediment texture.
- Bacterial activity can denote the extent of arsenic adsorption/desorption.

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure to arsenic in the environment is hazardous to biota. Three types of sediments from a shallow, high arsenic aquifer were collected from Jianghan Plain, Central China to investigate interactions between indigenous bacterial activity and arsenic adsorption/desorption. For the same level of bacterial activity, the As(III) or As(V) adsorption rate in sediments decreased from clay loam to loamy sand to silty sand. However, the arsenic desorption rate from these sediments followed the reverse sequence. For the same arsenic speciation and content, bacterial activity decreased from clay loam to loamy sand to silty sand. Overall, arsenic adsorption/desorption rates were related to both bacterial activity and sediment texture. The bacteria present could reduce As(V) to As(III), thereby favoring its release from sediment into solution and increasing the ratio of As(III) to total arsenic. These results indicate indigenous bacteria strongly affect the adsorption/desorption and oxidation-reduction of arsenic, and are actively involved in the dynamic equilibrium of arsenic between sediment and groundwater in this shallow aquifer.

1. Introduction

Arsenic is a ubiquitous toxic metalloid, and elevated levels of arsenic in soil and groundwater is a serious environmental problem (Jiang et al.,

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2013). Arsenic-contaminated drinking water results in adverse health effects and long-term exposure can lead to arsenicosis (Fan et al., 2018; Nguyen et al., 2011; Oremland and Stolz, 2005).

Arsenic is found in the subsoil incorporated in >245 minerals, but mostly as metal sulfides (Álvarez-Benedí et al., 2005). Elevated arsenic levels in the environment can be derived from both natural and anthropogenic inputs (Alexakis, 2011; Kelepertsis et al., 2006). Enrichment of As in sediments can also be associated with geothermal activity (Alexakis and Gamvroula, 2014). Arsenic trioxide converted from arsenic sulfides in rocks is released into the arsenic cycle (Mandal and Suzuki, 2002). Arsenic can occur in many different physicochemical forms, but in terrestrial and aquatic environments is predominantly present as inorganic arsenite [As(III)] and arsenate [As(V)] (Ohtsuka et al., 2013). Organic arsenic, which dominates in biomass, has also been detected in soil (Leonard, 1991). Forms of organic arsenic include monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), and arsenobetaine (Ko et al., 2004). These organic forms of arsenic are less toxic than inorganic forms (Xie et al., 2013a). Microbes are responsible for many reactions that result in the transformation of arsenic species. Certain microbes can biomethylate inorganic forms to gaseous arsines or MMA and DMA, while other microbes can demethylate organic forms to inorganic species (Sohrin et al., 1997). Based on its speciation and chemical characteristics, arsenic can enter the food chain where it becomes widely distributed in both plants and animals (Mandal and Suzuki, 2002; Ruíz-Huerta et al., 2017). Adsorption/desorption processes play an important role in arsenic entering the food chain. Considerable efforts have been made to study the adsorption/desorption of arsenic, but the major focus has been on abiotic factors, such as pH, redox conditions, solution composition, and mineral substrate composition (Bowell, 1994; Haque et al., 2008; Kuhn and Sigg, 1993; Maest et al., 1992). Phosphates significantly decrease the adsorption of arsenic (Álvarez-Benedí et al., 2005), while nitrate shows the opposite effect. Small substrate particles adsorb more arsenic than large particles (Caporale et al., 2013).

Redox changes can take place in groundwater environments and affect the forms of inorganic arsenic present. The reduction of As(V) to As(III) is a very slow process under naturally reducing conditions. Hence, As(V) is the predominant species absorbed by sediments; this affects the mobilization and transformation of arsenic species (Álvarez-Benedí et al., 2005) and is relevant to consider in addition to As(III) being much more toxic and mobile than As(V) in groundwater systems (Oremland and Stolz, 2005). Therefore, the mobilization and transformation of arsenic is related to both redox conditions and adsorption/desorption processes. Many recent studies have also documented the important role of microbes in arsenic migration and speciation change (Ma et al., 2016; Oremland et al., 2005; Park et al., 2006; Turpeinen et al., 1999; Xie et al., 2011).

Hence, determining the role of microbes as well as adsorption/desorption and oxidation/reduction is very important for predicting the environmental effects of arsenic. The major objectives of this study were: (i) to determine microbial oxidation/reduction of arsenic; (ii) to explore the relationship between oxidation/reduction and adsorption/desorption in arsenic migration; and (iii) to evaluate the effects of the indigenous bacterial growth on arsenic adsorption/desorption processes and its response in a shallow, high arsenic groundwater system located in the Jianghan Plain, Central China. The theoretical conclusions drawn from the results are important for better understanding microbial effects on arsenic accumulation in soil and shallow groundwater systems.

2. Materials and method

2.1. Site description

Jianghan Plain, located in central China, is a major crop production area for China and is known as 'a land of milk and honey'. The sampling site (30°09′ N, 113°41′E) was in Shahu town, Xiantao city, Hubei province, situated between the Yangtze River and the Han River. The average annual precipitation is 1115 mm, with maximum and minimum temperatures of 43 °C (July) and −10 °C (January), respectively.

Due to surface water contamination resulting from intense human activities, groundwater has increasingly been used as a main source of drinking and irrigation water for the Jianghan Plain. However, groundwater arsenic concentration is up to 175 μg L^{-1}, resulting in many people being diagnosed with arsenic poisoning in this area (Chen et al., 2007).

2.2. Sediment sampling and characterization

Based on previous sampling surveys for arsenic content and sediment properties, three sediments (S1, S2 and S3) were collected in November 2011 from a 50-m deep borehole in the study area. Sampling depths for S1, S2, and S3 were 11.5–11.8, 13.2–13.5 and 14.3–14.6 m, respectively. Each 30-cm core sample was in situ packed in polyethylene bags, sealed in a PVC pipe immediately after retrieval, and then stored in a refrigerator at 4 °C. Samples were delivered to the laboratory as soon as possible, where the surface of the cores was removed and the remainder passed through an 11.2-mm sieve and air-dried at ambient temperature for 10 d. Dried sediments were crushed and passed through a 2-mm sieve. Sample textures were divided into sand (50–2000 μm), silt (2–50 μm), and clay (<2 μm) fractions.

Physico-chemical parameters including pH, total organic carbon (TOC), and concentrations of major and trace elements were determined. Sediment samples were added into deionized water at a ratio of 1:2.5 (wt/wt, sediment/water), and mixed with a magnetic stirrer. The pH of the sediment samples was then measured by a calibrated pH meter (PHS-3S, Shanghai Precision & Scientific Instrument Co., Ltd., China). The TOC of sediment samples was determined according to Yeomans and Bremner (1988). The method developed for the determination of several major and trace elements was based on the digestion of a 0.5-g sample of dry sediment with HNO_3/HCl (8 and 2 mL, respectively) in a microwave oven at 180 °C for 20 min. The solution was then evaporated to dryness and extracted with 20 mL of diethylene triamine pentaacetate. The mixture was then analyzed for several major and trace elements by inductively coupled plasma atomic emission spectrometry (ICP-AES) (IRIS Intrepid II XSP, Thermo Elemental). The As(III) and As(V) content in the sediment were determined according to Shi et al. (2003), with detection limits for both of 0.01 mg kg^{-1}.

Determination of the number of arsenic resistant bacteria in the three types of sediments followed the method of Corsini et al. (2010) with slight modifications. About 5 g of fresh sediment sample was suspended in 45 mL of sterile distilled water in a 250-mL conical flask, shaken on a rotary shaker at 150 rpm for 2 h, and then left to settle for 10 min. Aliquots (1 mL) of the sediment suspension were subjected to a 10-fold serial dilution in 0.9% NaCl solution. Subsequently, 0.1 mL of the serial dilutions was spread on 1:10 diluted Tryptic Soy Agar medium with 1 mM As(III) or As(V). The number of As-resistant bacteria was calculated according to the dilution of the suspension and number of colony forming units (CFU) on the plate.

2.3. General methods for assays

Stock arsenic solutions containing 10 mM As(III) ($NaAsO_2$) or As(V) ($Na_3AsO_4 \cdot 12H_2O$) were prepared with deionized water (Super-Q plus, Millipore Co., Ltd., US) at the beginning of the experiment.

All glassware and plasticware was soaked in 5% (v/v) HCl solution for at least 24 h, followed by repeated rinsing with deionized water before use. All chemicals were analytical reagent grade and all reagent solutions were prepared with deionized water. Oxygen-free experimental conditions were achieved through purging with high-purity N_2 for at least 1 h. Sediment results are presented on a dry weight basis.