



Effect of reductive dissolution of iron (hydr)oxides on arsenic behavior in a water–sediment system: First release, then adsorption



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ABSTRACT

Whether the reductive dissolution of iron (hydr)oxides promotes absorption or desorption of arsenic (As) is still in controversy. Here, we used laboratory simulation experiments to examine the absorption/desorption behavior of As when it is affected by microbial activity. Sediment from Yangzonghai Lake, which is known to be contaminated by As, was used in this experiment, and pollution abatement was performed using ferric salt flocculants (FeCl₃). In the presence of sulfides, the reductive dissolution of iron (hydr)oxides affects the behavior of As in sediment in two ways. First, the reductive dissolution of iron (hydr)oxide accelerates the desorption of As, thus releasing As(III) from the sediment, increasing the environmental risk of As. Secondly, reductive dissolution causes the system's redox conditions to continually change. When the oxidation–reduction potential (Eh) of the system exceeds 80 mV, As is precipitated rather than desorbing. As is co-precipitated with sulfur released previously via microbially induced reductive dissolution, where sulfur precipitation is more important than reductive dissolution of iron (hydr)oxide in controlling the adsorption/desorption of As.

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1. Introduction

In recent years, arsenic (As) contamination of water bodies has posed a serious problem internationally (Baig et al., 2013; Freikowski et al., 2013). It is believed that As contamination of drinking water poses a significant cancer risk at concentrations >50 μg L⁻¹ (Rahman et al., 2001). According to a previous report, the As concentration in the drinking water for the 30 million people who reside in the western part of the Bengal Delta Plain exceeds 50 μg L⁻¹ (Freikowski et al., 2013). China also must contend with As contamination; previous studies have estimated that about 20 million people live in areas at risk of As contamination, including water bodies in the Xinjiang, Inner Mongolia, Shanxi, Hunan, Yunnan, Guizhou, Guangdong, Guangxi, and Taiwan provinces (Rodríguez-Lado et al., 2013). At present, As contamination in drinking water sources threatens the health of some 200 million people worldwide (Baig et al., 2013).

In aquatic ecosystem restoration, iron chloride (FeCl₃) is widely used for improving water clarification or used as a flocculant to remove contaminants such as phosphorus, Cu, Pb, Zn and arsenic (As) from water (Contin et al., 2015; Kim et al., 2014) by

co-precipitation of both constituents as contaminant-containing iron (oxyhydr)oxides (Zaaboub et al., 2014), and transfer contaminants from water into sediments. However, some researches showed that iron (hydr)oxides precipitated during sewage sludge flocculation were partially redissolved the acid soil, releasing previously surface adsorbed contaminants (Contin et al., 2015). Yangzonghai Lake, which was contaminated by As and then treated with FeCl₃ (Liu et al., 2014) during September 2009–December 2011, resulting in a large quantity of insoluble complex as As–iron (hydr)oxides in sediment. The changes of redox potentials (Eh) in the overlying water may lead to dissolve of the iron (hydr)oxides, whether the reduction dissolve promotes the adsorption/desorption of As should be investigated further.

Arsenic exists mainly in the form of As(III) and As(V) in water environments, in which the toxicity and mobility of As(III) are greater than those of As(V). After entering water environments, As undergoes geochemical processes such as adsorption/desorption, precipitation or co-precipitation, and oxidation/reduction (Smedley and Kinniburgh, 2002; Xu et al., 2011). These processes change the form of As within the environment, thus resulting in fixation or migration because different forms of As have different stabilities in water (Burton et al., 2011). However, the main factors that control the form of As in water and solid phases remain unclear. Many factors influence the migration and transformation of As; for example, hydrodynamics, pH–Eh, competing ions,

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organic matter, and microbes. Because As has a high affinity for iron (hydr)oxides (Lowers et al., 2007), much attention has been paid to how Fe reduction increases the concentration of As in groundwater. Previous studies have shown that the reductive dissolution of Fe, with the participation of microbes, plays an important role in managing the migration and transformation of As (Smedley and Kinniburgh, 2002; Islam et al., 2004). It is generally thought that the microbially mediated reductive dissolution of iron-bearing minerals in water environments leads to the release of As. The amount released depends on the valence state of As and the mineralogical characteristics of Fe within the environment (Islam et al., 2004; Campbell et al., 2006). However, some studies indicate that the reductive dissolution of As bound to iron (hydr)oxides can be ignored (Kocar et al., 2006), because the reductive dissolution of iron (hydr)oxides promotes the adsorption rather than desorption of As (Tufano and Fendorf, 2008). Consequently, controversy remains concerning whether the reductive dissolution of iron (hydr)oxides in sediment would cause adsorption or desorption of As.

Microbes can reduce sulfates, which will then decrease the solubility of As bound to sulfides and precipitates (Camacho et al., 2011). Sulfides play an important role in As dynamics within aquatic environments (Xu et al., 2011). This makes determining the fate, form and concentration of As within these complex environments difficult. Currently, how sulfide ions participate in the secondary fixation of As remains unclear.

Iron-based materials have been developed to reduce the As concentration in drinking water that has As below the World Health Organization limit of $10 \mu\text{g L}^{-1}$ (Corsini et al., 2014). In this study, we selected the Yangzonghai Lake as an example of a deep lake the As contamination of which had been treated by spraying FeCl_3 . We designed the sediment–water interface simulation experiments so as to provide some scientific data for the ecological risk assessment of ferric salt treatment in As contaminated lake water restoration engineering. Our laboratory experiments aimed to investigate: (1) the influence of the reductive dissolution of iron (hydr)oxides on As in the presence of Fe-rich sediments and

enhanced microbial activity, and (2) the adsorption/desorption mechanism of As in the presence of Fe and S in sediment, as well as the presence of active anaerobes.

2. Material and methods

2.1. Study region overview

Yangzonghai Lake is a fresh water lake located in southwest China ($24^{\circ}51'–24^{\circ}58'N$, $102^{\circ}59'–103^{\circ}02'E$). This lake is also the main water supply for agriculture and fisheries in the area, as well as an alternate drinking water source for the local community. The lake is located at an elevation of 1770 m, and has a maximum depth of 30 m and an average depth of 20 m. This lake has a catchment area of 192 km^2 , with an average annual water storage of $5.69 \times 10^8 \text{ m}^3$. For the Yangzonghai Lake, the average annual water temperature is 18°C . In 2008 the lake experienced an accidental As contamination; because of this, As concentrations peaked at $134 \mu\text{g L}^{-1}$ in October of that year (Liu et al., 2014), when all human uses of the lake water ceased. Subsequently, attempts to abate the concentrations of As were attempted by spraying FeCl_3 . These efforts decreased the concentration of As below $50 \mu\text{g L}^{-1}$, according to a measurement taken in August 2011. Such Fe salts are well suited for reducing the concentration of As within water bodies, as they form insoluble complex compounds with As that precipitate to the bottom of the lake (Liu et al., 2014). Yangzonghai Lake is a deep lake, with thermal stratification in the summer leading to strong anaerobic conditions at the bottom of the lake (Liu et al., 2014). Therefore, investigating whether the reduction dissolution of iron (hydr)oxides under anaerobic conditions causes the adsorption or desorption of As is critical for evaluating the risk of secondary pollution in the lake.

2.2. Sampling and cultivation of anaerobic microbes

Samples were collected on November 5, 2013. Thirty-one sampling sites were selected in the lake using the rectangular grid

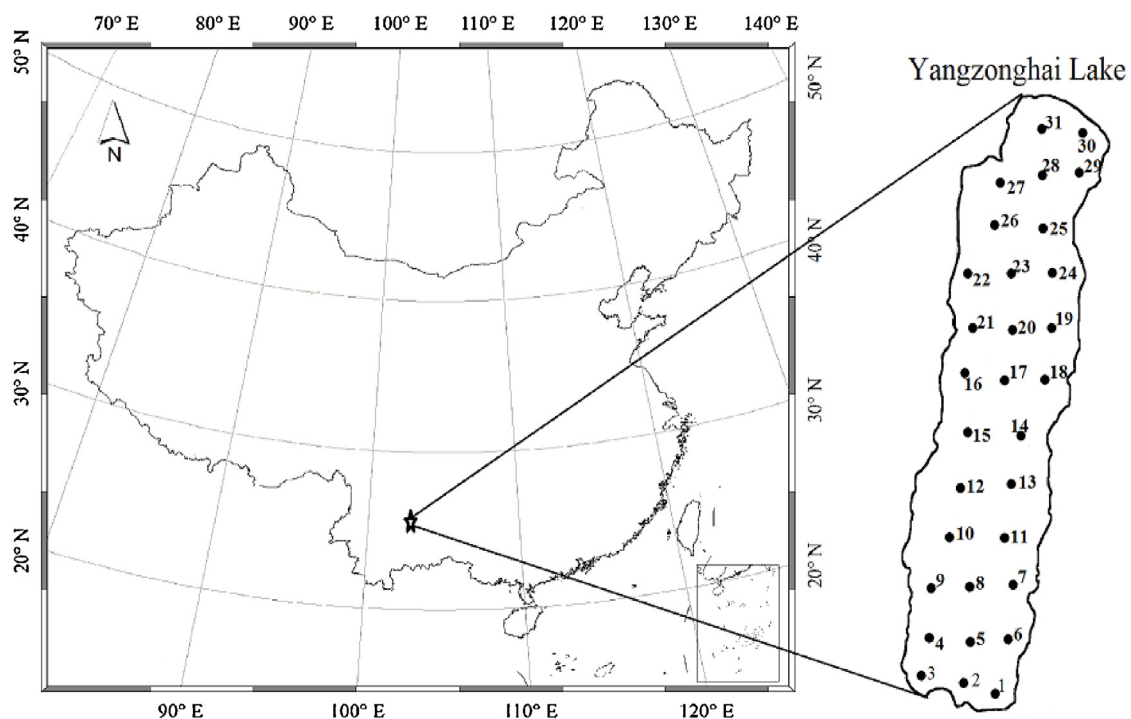


Fig. 1. Schematic of sampling sites in Yangzonghai Lake in southwest China.

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