



Effects of elevated sulfate concentration on the mobility of arsenic in the sediment–water interface

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ARTICLE INFO

Keywords:

Arsenic contamination
Lake sediment
Sediment–water interface
Water pollution

ABSTRACT

The adsorption/desorption of arsenic (As) at the sediment–water interface in lakes is the key to understanding whether As can enter the ecosystem and participate in material circulation. In this study, the concentrations of As(III), total arsenic [As(T)], sulfide, iron (Fe), and dissolved organic carbon (DOC) in overlying water were observed after the initial sulfate (SO_4^{2-}) concentrations were increased by four gradients in the presence and absence of microbial systems. The results indicate that increased SO_4^{2-} concentrations in overlying water triggered As desorption from sediments. Approximately 10% of the desorbed As was desorbed directly as arsenite or arsenate by competitive adsorption sites on the iron salt surface; 21% was due to the reduction of iron (hydr)oxides; and 69% was due to microbial activity, as compared with a system with no microbial activity. The intensity of microbial activity was controlled by the SO_4^{2-} and DOC concentrations in the overlying water. In anaerobic systems, which had SO_4^{2-} and DOC concentrations higher than 47 and 7 mg/L, respectively, microbial activity was promoted by SO_4^{2-} and DOC; As(III) was desorbed under these indoor simulation conditions. When either the SO_4^{2-} or DOC concentration was lower than its respective threshold of 47 or 7 mg/L, or when either of these indices was below its concentration limit, it was difficult for microorganisms to use SO_4^{2-} and DOC to enhance their own activities. Therefore, conditions were insufficient for As desorption. The migration of As in lake sediments was dominated by microbial activity, which was co-limited by SO_4^{2-} and DOC. The concentrations of SO_4^{2-} and DOC in the overlying water are thus important for the prevention and control of As pollution in lakes. We recommend controlling SO_4^{2-} and DOC concentrations as a method for controlling As inner-source pollution in lake water.

1. Introduction

Arsenic (As) is a carcinogen. In recent years, As pollution incidents have increased the concentration of As in natural water, thereby threatening human health (Bhattacharya et al., 2007; Bundschuh et al., 2013). Mining (Rieuwerts et al., 2014; Gallego et al., 2015) and industrial emissions are important sources of As pollution (Liu et al., 2014; Gallego et al., 2015). The prevention and treatment of As pollution has become a focus of research and discussion (Meliker et al., 2007; Bhattacharya et al., 2007; Marabottini et al., 2013). Understanding the ecotoxicological effects of As in the environment is important for mitigating its deleterious environmental and human health effects (Bundschuh et al., 2013), and remediating As-contaminated

water supplies is important (Bhattacharya et al., 2007). In natural fresh water, arsenate (AsO_4^{3-}) and arsenite (AsO_3^{3-}) are the two major forms of As (Cullen and Reimer, 1989), and these can be precipitated with iron (hydr)oxides (Root et al., 2007; Chen et al., 2015). Therefore, iron salt flocculants are commonly used to treat As-polluted water based on the principle of adsorption precipitation (Root et al., 2007; Chen et al., 2015), and these As–iron complexes then settle into the sediment at the bottom of the lake. The As can be released into solution through the reduction of iron (hydr)oxides (Pedersen et al., 2006; Vink et al., 2017), possibly facilitated by the metabolic activity of specific microbial populations (Macur et al., 2004), or it may desorb when other environmental conditions in the overlying water change (Smedley and Kinniburgh, 2002; Xu et al., 2011; Burton et al., 2011; Liu et al., 2014),

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resulting in secondary As pollution of the lake water.

The sediments in As-contaminated areas usually have elevated As concentrations (Rieuwerts et al., 2014). The sulfate (SO_4^{2-}) concentration in water increases in environments with long-term acid deposition. Sulfate can be adsorbed on minerals as outer or inner complexes (Zhang and Sparks, 1990; Kinjo and Pratt, 1971), and competes with AsO_4^{3-} or AsO_3^{3-} to occupy adsorption sites on iron (hydr)oxides. This process reduces the amount of As adsorbed on mineral surfaces and promotes the dissolution of As in sediments (Xu et al., 1988; Manning and Goldberg, 1996a, 1996b; Song et al., 2015). In addition, SO_4^{2-} can also be used by microorganisms to further promote the release of As. The sulfate-reducing microorganisms in the sediments promote the reduction of iron (hydrogen) oxides of ferric iron to ferrous iron compounds like FeS and FeS_2 . These iron–sulfur compounds have a weak fixation capacity for As. As a result, the As, previously adsorbed on the iron (hydrogen) oxide, is desorbed and migrates to the water (Kocar et al., 2010; Burton et al., 2011). AsO_3^{3-} is thus released from the surface of the metal salt due to the action of microorganisms. It is unclear whether the increased SO_4^{2-} concentration in overlying water caused by acid deposition affects As mobilization by ion competition or by stimulating microbial activity and its mechanism of impact.

Yangzonghai Lake is located in southwest China (24°51′–24°58′ N, 102°59′–103°02′ E, Yang et al., 2015). In June 2008, the lake suffered a serious As pollution incident, and the maximum concentration of As reached 0.128 mg/L in September (Chen et al., 2015). Iron salt flocculants (FeCl_3) were chosen to treat the As pollution in the lake. The mechanism involves the adsorption of As to amorphous $\text{Fe}(\text{OH})_3$ produced by the hydrolysis of FeCl_3 , and subsequent precipitation (Chen et al., 2015). The treatment lasted 2 years, from October 2009 to November 2011. At the beginning of the treatment period, 6–8 t of FeCl_3 flocculants were sprayed into the lake daily (Chen et al., 2015). Considerable amounts of high-iron and high-arsenic sediments were formed during the treatment process and stored at the bottom of the lake. The flocculants in these sediments were not saturated (Liu et al., 2014). The lake also suffers from long-term acid deposition (Wang et al., 2005). In the 1950s, Yangzonghai Lake had good water quality. In the early 1960s, it was impacted by atmospheric deposition of coal and smoke. When the pollution intensity peaked from 1965 to 1980, the minimum instantaneous pH of the lake water was reduced to 5–6. From 1983–1999, the pollution intensity tended to decrease, but it remained a major obstacle to environmental remediation, and acid deposition led to the spread of acid-tolerant species and the demise of shellfish (Wang et al., 2005). The impact of changes in SO_4^{2-} anion concentrations in overlying water on the adsorption/desorption of high-iron and high-arsenic sediments is unclear. In this study, four SO_4^{2-} concentration gradients were designed to simulate increasing SO_4^{2-} concentrations in overlying water, and to determine when the increase in SO_4^{2-} concentration occurred. Specifically, the study examined: (1) whether increased SO_4^{2-} concentrations induce As desorption/adsorption; (2) the mechanism of As desorption/adsorption, i.e., whether As is desorbed by competitive anion exchange with arsenate/arsenite, by microbial action, or through the reduction of iron (hydr)oxides in the presence of excess Fe. Our results will improve risk assessment for water contamination resulting from the large-scale storage of As in lake sediments.

2. Materials and methods

2.1. Sampling

Samples were collected from Yangzonghai Lake on May 5, 2015. FeCl_3 flocculants were used to treat As pollution in the lake between 2009 and 2011 (Chen et al., 2015). The lake was previously described by Liu et al. (2014) and Yang et al. (2015). Surface sediment samples were collected from the south of Yangzonghai Lake using an Ekman B08 bottom grab sampler (Ekman, Beijing, China). The depth of the water at

the sampling sites was 22 m. Before sediment samples were collected, approximately 125 L of overlying water was collected using a pump. These water samples were stored in 25-L plastic buckets. Each sediment sample was stored in a sealed sterile sample bag to ensure that an anaerobic environment was maintained. Samples were stored at 0–4 °C. After collection, wet samples were transported to the laboratory, where they were thoroughly mixed in a glove box under anaerobic conditions.

2.2. Experimental simulation

2.2.1. Sampling and experimental design

Both the sediment and water samples from Yangzonghai Lake were used in the experimental simulation using a water-to-sediment mass ratio of 20:1 (4000 mL water, 200 g sediment). The As concentration of the overlying water was adjusted to 0.128 $\mu\text{g/L}$ (i.e., the As concentration at the time of the 2008 As pollution incident in Yangzonghai Lake) through the addition of sodium arsenite. Experiments were conducted under two conditions: non-microbial conditions and microbial conditions.

In the experiment with non-microbial conditions, both the sediment and water samples were sterilized. The water and sediment samples were placed in sterile barrels and glass containers, respectively, and then placed in a high-pressure microbial pot (Shanghai Shen An Instrumentaria, DSX-280B) at 121 °C for 30 min (Xu et al., 2011), three times. Subsequently, 24 200-g aliquots of the sterilized wet sediment were weighed and placed in separate glass sample tanks. Sterilized water (4 L) was slowly added to each tank along the tank wall on a sterile console after cooling. FeCl_3 flocculants were then added to the tanks at a concentration of 0.1 g/L. The water was gently stirred to simulate the flocculent treatment of As contamination. The As concentration in the overlying water of each group was subsequently decreased to less than 10 $\mu\text{g/L}$. After treatment, the concentration of As in the overlying water of each tank was determined. Sediments containing high As and high iron concentrations were thus obtained. Na_2SO_4 was added on the third day to regulate the SO_4^{2-} ion concentration of the overlying water in four gradients (six replicates per gradient): 54 (no addition, maintaining the initial concentration), 110, 329, and 547 mg/L; these sterilized groups were designated S_0 (no addition group, used as a control), S_1 , S_2 , and S_3 , respectively. Next, we used nitrogen (N_2) gas to displace oxygen (O_2) from the tanks. The dissolved oxygen (DO) concentration was determined at this time. When the DO level decreased to below 3.0 mg/L, we terminated the N_2 purge. The tanks were then sealed and covered with dark cloth.

In the experiment with microbial conditions, the treatments were consistent with the experiment with non-microbial conditions, but the samples were not sterilized. Four gradients were used in the groups with microbial conditions: 54, 110, 329, and 547 mg/L. They were designated M_0 (no addition group, used as a control), M_1 , M_2 , and M_3 , respectively.

The overlying water samples of all experimental groups in the simulation experiment were taken on days 1, 4, 7, 10, 17, 24, 31, 38, 45, 52, 67, and 97 after the start of the experiment. The pH and DO of the overlying water were monitored at the sampling time. All samples were taken to determine the concentrations of Fe, total As [As(T)], As(III), dissolved organic carbon (DOC), SO_4^{2-} , and S^{2-} . The volume of aqueous samples was 10 mL in all cases, and all samples were centrifuged (954 × g, 10 min) to remove solid particles from the solution.

2.2.2. Measurements

The pH of the system was evaluated using a HI8424 NEW Portable pH/mV/1 C Meter (Hanna Instruments, Padova, Italy) and the DO concentration was determined using a DO meter (Hanna Instruments). The supernatant was decanted and subsampled for further analysis. As (T) and As(III) concentrations were determined using an AFS-933 at fluorescence spectrometer (Titan Instruments, Beijing, China; detection limit = 0.5 $\mu\text{g/L}$). To determine As(T), the samples were pretreated

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