



# First observation of labile arsenic stratification in aluminum sulfate-amended sediments using high resolution Zr-oxide DGT



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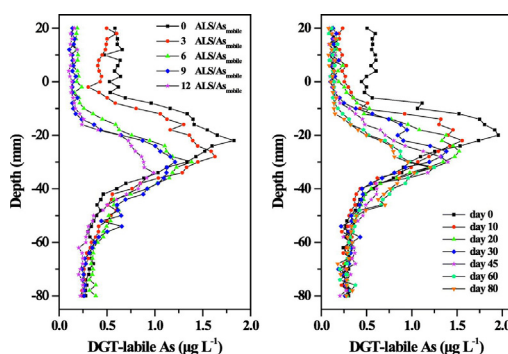
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## HIGHLIGHTS

- Labile As in alum-amended sediments was first investigated on mm scale.
- Concentration of labile As decreased with increasing dosage and time.
- A static layer appeared with low labile As and variation in the top 12 mm sediment.
- Upward diffusion of labile As was reduced below static layer with relaxed gradient.
- As release in sediments below static layer was minimized by increasing  $K_d$  and  $k_1$ .

## GRAPHICAL ABSTRACT



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## ABSTRACT

Arsenic contamination in sediments has received increasing attention because it may be released to the water and threaten aquatic organisms. In this study, aluminum sulfate (ALS) was used to immobilize As in sediments through dosage-series and time-series experiments. Diffusive gradients in thin films (DGT) was used to obtain labile As at a vertically 2.0 mm resolution. Our results indicated that a “static” layer with extremely low labile As concentration (minimally  $0.13 \text{ mg L}^{-1}$ ) with weak variation ( $<30\%$  RSD) formed within the top 12 mm sediment layer at the dosage of 6–12  $\text{ALS}/\text{As}_{\text{mobile}}$  ( $\text{kmol mol}^{-1}$ ,  $\text{As}_{\text{mobile}}$  means the total mobile As in top 40 mm sediment) and on days 30–80 after amendment at the dosage of 9  $\text{ALS}/\text{As}_{\text{mobile}}$ . The maximum labile As decreased from  $1.83$  to  $0.99 \mu\text{g L}^{-1}$  and from  $1.96$  to  $1.20 \mu\text{g L}^{-1}$  in the dosage-series (3–12  $\text{ALS}/\text{As}_{\text{mobile}}$ ) and time-series (10–80 days) experiments, respectively, while the depths showing the maximal concentrations moved deeper from 22 to 34 mm and 20 to 32 mm in the sediments. It implied a reduced upward diffusion potential of labile As to the static layer in deeper sediments. Both distribution coefficient for As between sediment solid pool and pore water ( $K_d$ ) and the adsorption rate constant ( $k_1$ ) consistently increased, reflecting that As release from sediment solid became increasingly difficult with the progress of ALS immobilization. The results of this millimeter-scale investigation showed that ALS could efficiently immobilize As in sediments under simulated conditions.

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## 1. Introduction

Arsenic (As) is a ubiquitous element in the environment, and it is one of the most hazardous chemicals in the world (Smedley and Kinniburgh, 2002). Inorganic arsenic species are well-known environmental toxicants and carcinogens. They can have significant health impacts on both humans and animals, and can lead to skin, liver, lung, kidney, and bladder cancers (Ng, 2005; Smith et al., 1992). Natural geological sources are the main causes of As contamination in drinking water around the world (Sharma and Sohn, 2009). However, human activities, such as mining, fossil fuel combustion, and the use of arsenic-containing products, have important impacts on As contamination (Smedley and Kinniburgh, 2002). Because of its toxicity and widespread occurrence, As contamination has become a serious problem worldwide and has attracted increasing attention (Du et al., 2015). Several physicochemical techniques have been used for As removal, including conventional treatment processes such as coagulation-precipitation, adsorption, ion exchange, and membrane processes (Mondal et al., 2013), and advanced processes such as reverse osmosis (Ning, 2002), electrodialysis (Sharma and Sohn, 2009), and electrocoagulation (Nidheesh and Singh, 2017). Arsenic species can also be removed using biological treatments like phytoremediation, biological filtration, and microbial remediation (Sarkar and Paul, 2016). Most of these techniques have focused on water bodies including wastewater, drinking water, and groundwater.

Sediments can accumulate a significant quantity of As, which arouses increasing concerns about As contamination in sediments (Nikolaidis et al., 2004). Sedimentary As may significantly influence lake water depending on the prevailing physicochemical conditions of the sediment-water interface (SWI) (Gorny et al., 2015). High As levels in sediments can be released through desorption and dissolution, endangering benthos and aquatic organisms (Zan et al., 2014). To date, however, there have been limited studies on immobilizing As in sediments. Ferrous materials ( $\text{FeCl}_3$  and  $\text{Fe}(\text{OH})_3$ ) were applied to immobilize As in evenly-mixed surface sediments, while  $\text{FeCl}_3$  significantly reduced the exchangeable As and changed the As species in the sediments (Huang et al., 2014). Mineral amendment (5% hematite) was used to immobilize As in leachate of contaminated harbor sediments and it reduced arsenic mobility by 30–40% (Mamindy-Pajany et al., 2010). Lime has been proven to be effective in reducing As mobility via the formation of low-solubility calcium arsenates and has been used in slurries with various Ca/As ratios (Bothe and Brown, 1999).

The existing studies on As immobilization concern the overall effectiveness of chemical agents on the mixed surface sediments or the leachates, but do not consider the heterogeneous characters of the sediments. Previous studies have shown significant vertical variations in the distributions of labile As concentration in sediments even on a millimeter scale (Sun et al., 2016b; Wang et al., 2016a; Yao et al., 2016). This is due to the reductive dissolution of Fe-bound As and the conversion of As(V) from its strongly adsorbed form into more weakly adsorbed form driven by the transition of redox condition near the SWI (Yao et al., 2016). Moreover, capping/mixing with immobilizing reagents, the adsorption of pore water and weakly adsorbed As by the reagents induces the upward diffusion of pore water As from deeper layer and intensifies the vertical heterogeneity in the labile As distribution in sediments. High-resolution methods for measuring the in-situ distributions of As in sediments are highly required. At present, Zr-oxide diffusive gradients in thin films (Zr-oxide DGT), a passive sampling technique, has been developed to satisfy the in situ measurement of As on a millimeter scale (Sun et al., 2014; Zhang et al., 2014). This technique is established based on Fick's first law. It measures the pore water As plus the easily-exchangeable As dynamically released from sediment solids to resupply the pore water As.

Iron and Al salts are commonly used reagents in coagulation-precipitation processes for As removal from water and wastewater because of their availability, low cost, and high removal efficiencies (the

ratio of removed mass of As to its initial mass in water or wastewater) (Hu et al., 2012; Pramanik et al., 2016). Iron salts are usually more promising for arsenic adsorption from water because they have higher removal efficiency than other adsorbents (Khan et al., 2002). However, the Fe compounds may transform into reduced forms and cause the reductive dissolution of arsenic iron compounds, which may risk mobilizing As in sediments (Bauer and Blodau, 2006; Chen et al., 2016b; Sun et al., 2016a). The Al-based salts, such as polyaluminum chloride (PACl),  $\text{AlCl}_3$ ,  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , and alum, are widely used to remove As from water, and the reported removal efficiencies vary widely from 40%–100% under different conditions (Hu et al., 2012; Lin et al., 2015; Pramanik et al., 2016). To date there are few reports about the uses of Al-based salts in immobilization of As in sediments.

In this study, we tested aluminum sulfate as the coagulant in simulated experiments for As immobilization in sediments. The Zr-oxide DGT was used to measure labile As in sediments at a vertical resolution of 2.0 mm. Dosage-series and time-series experiments were performed to investigate the changes in the vertical distribution of labile As after ALS amendments. The DGT Induced Fluxes in Sediments (DIFS) model was used to simulate the solute resupply kinetics during the progress of ALS immobilization.

## 2. Materials and methods

### 2.1. Sample collection

Water and surface sediment samples were collected from the Nanfei River, which is one of the eight major inflowing rivers for Chaohu Lake (Hefei City, Anhui Province, China). The Nanfei River is a heavily polluted river due to the wastewater discharge of heavy polluting industries in Hefei City (Shao et al., 2016).

Sediment was collected with a Peterson Grab (length  $\times$  width  $\times$  height =  $36 \times 20 \times 15$  cm) at the sampling site. After being transferred to the laboratory, the water sample was filtered using a  $0.45 \mu\text{m}$  cellulose acetate membrane filter, and the wet sediment sample was sieved using a  $0.6$  mm pore-sized mesh to remove coarse particles. The sediment was homogenized and put into Perspex tubes ( $8.4$  cm in internal diameter,  $33$  cm in length) with a depth of about  $15$  cm, and then the lake water was siphoned into the tubes to a depth of about  $12$  cm. The microcosms were pre-incubated in a water bath at  $25^\circ\text{C}$  for  $60$  days. After pre-incubation, three sediment cores were retrieved and the sediments in the upper  $40$  mm layer were used for measurement of  $\text{As}_{\text{mobile}}$  using the method described below.

### 2.2. Experimental microcosm set-up

#### 2.2.1. Principle and preparation of DGT probes

The DGT device consists of a binding layer (binding gel) and a diffusion layer (diffusion gel plus a filter membrane). The soluble element diffuses through the diffusion layer and gets quickly bound in the binding layer, leading to the formation of a linear diffusion gradient, across the diffusion layer (Zhang et al., 2014). Based on this gradient, DGT can measure free ions and labile complexes in solution that can dissociate in the time required to traverse the diffusive layer (Warnken et al., 2008). In sediments, DGT uptake of soluble fractions in pore waters leads to the dynamic release of weakly-bound fractions to replenish the pore water concentration. Hence, DGT measurement reflects the concentrations of determinants in pore water and labile solid pool as well as the characteristics of their dynamic interactions (Zhang et al., 1995).

The Zr-oxide DGT probes (Sun et al., 2015) are used for the measurement of labile As, which were provided by EasySensor Ltd. ([www.easysensor.net](http://www.easysensor.net)). The diffusion gel and binding gel were prepared according to Wang et al. (2016b) and Xu et al. (2013), respectively. To assemble the DGT probe, the Zr-oxide binding gel was put on the base of a new flat DGT holder and covered with the agarose diffusive gel and a

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