



A millimeter-scale observation of the competitive effect of phosphate on promotion of arsenic mobilization in sediments



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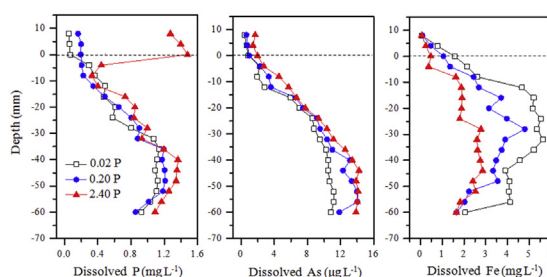
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HIGHLIGHTS

- The competitive effect of P on As release in sediments was first investigated on a mm scale.
- Elevation of water P increased DGT-labile P and As but it decreased DGT-labile Fe.
- Limited increases in dissolved P and As appeared due to sediment buffering.
- Resupply ability of sediment solids for pore water As increased with elevation of water P.
- Fe(II) precipitation greatly suppressed P competition on As mobilization under low P treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

A millimeter-scale investigation is key to the understanding of the competitive effects of phosphate(P) on arsenic(As) mobility in sediments by taking the great biogeochemical heterogeneity of the sediments into consideration. In this study, a microcosm experiment was performed in this aspect using high-resolution dialysis and diffusive gradients in thin films (DGT) to simultaneously measure dissolved and labile P, As, and iron (Fe) in sediments, respectively. With the increase of P content in water from 0.02 mg L⁻¹ to 0.20 and 2.4 mg L⁻¹, consistent release of As from sediments was observed. The concentrations of DGT-labile As increased significantly especially in the upper sediment layer (up to 12 times of the 0.02 mg P L⁻¹ treatment) due to the competition of phosphate, which corresponded well to the increase in DGT-labile P. There was limited increase in dissolved P and As contents due to the buffering provided by sediment solids, while the concentrations of both dissolved and DGT-labile Fe in sediments decreased. A stoichiometric calculation showed that 47% and 8% of the added P were removed through Fe(II) precipitation for the 0.20 and 2.4 mg P L⁻¹ treatments, respectively, which greatly suppressed the release of As induced by P competition for the 0.20 mg P L⁻¹ treatment. The DGT-induced fluxes in sediments (DIFS) modeling showed an increase in solid resupply to pore water As from elevation of water P through the increases of the desorption rate constant from 5.4 to 31(× 10⁻⁷) s⁻¹ and the sorption rate constant from 1.8 to 22(× 10⁻⁴) s⁻¹.

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1. Introduction

Arsenic (As) has ubiquitous presence in the Earth's crust, with an upper crustal abundance of 4.8 mg kg^{-1} (Rudnick and Gao, 2003). It is recognized as one of the most dangerous environmental hazards due to its adverse impact on human health as well as its toxicity towards plants and animals (Kaur et al., 2011; Mandal and Suzuki, 2002). Its long-term exposure to human beings increases the risk of cancers and non-cancer diseases (Ng et al., 2003; Sabath, 2015). It has been estimated that >100 million people are at risk of drinking water with As concentrations above the safe and permissible limit of $10 \mu\text{g L}^{-1}$ as recommended by the World Health Organization (WHO) (Rahman et al., 2009).

Sediment is a major sink for As in surface water, where the predominant oxidation forms of As are pentavalent and trivalent, i.e. As(V) and As(III) (Gorny et al., 2015). In oxidizing environments, arsenic is usually present as adsorbed, co-precipitated and mineral arsenate through association with various oxides and clay minerals (Jia et al., 2007; Hafeznezami et al., 2016). In reducing environments, arsenic is present in the form of arsenic sulfide in addition to adsorbed and co-precipitated arsenite (O'Day et al., 2004). As(-I) minerals (e.g. arsenopyrite and thioarsenates) may also be present in sulfidic environments (Hudson-Edwards et al., 2005; Sun et al., 2016a). When the oxic condition in surface sediment changes to anoxic condition in deeper sediment, arsenic may be mobilized and released in aqueous phase from reductive dissolution of iron oxides and/or reduction of adsorbed and/or coprecipitated As(V) to As(III) with weaker affinity with minerals (Bose and Sharma, 2002; Cummings et al., 1999; Pedersen et al., 2006; Zobrist et al., 2000; Gorny et al., 2015).

In addition to the above mechanisms, As in sediment may also be mobilized due to competitive desorption by competing anions, especially phosphorus (P) (Appelo et al., 2002; Gao et al., 2011; Rubinos et al., 2011; Silva et al., 2012). This process has been considered as a basic mechanism leading to the release of As under an oxic condition (Smedley and Kinniburgh, 2002). Phosphate shares similar physiochemical properties with inorganic As (arsenate in particular), likely producing potentially competing effects on retaining As by sediments. Similar findings have been well documented in soil sciences. For example, much attention has been paid to the interactions between As and P adsorption on clay minerals, Fe/Al oxides/hydroxides, and intact soils (Bolan et al., 2013; Liu et al., 2001; Pigna et al., 2009; Violante and Pigna, 2002; Zeng et al., 2008). Both As and P occupy similar adsorption sites, thus the addition of P can compete with As for surface sites in soils and correspondingly increase the mobility and availability of As (Garmo et al., 2008; Jiao et al., 2012). Despite that much attention has been paid to the competition of P on As mobilization, only a few investigations are available in this field (Gao et al., 2011; Rubinos et al., 2011). Gao et al. (2011) performed a batch experiment and observed that As concentration in waters significantly increased with the increase of added P concentration in waters, with the concentration of As increased to $13.6 \mu\text{g L}^{-1}$ under the highest phosphate loading of 30 mg L^{-1} . Rubinos et al. (2011) performed a microcosm investigation on an As-rich river bed sediment. The addition of phosphate (1 mM) increased the amount of As released (between 2 and 8 times) within the entire pH range (3–10) and period (168 h) tested. The desorption of As was considered to play a key role from the competition of P.

It has been increasingly recognized that the sediment has a great heterogeneity feature in biogeochemistry (Stockdale et al., 2009; Pagès et al., 2011; Ding et al., 2015; Santner et al., 2015). Correspondingly, the distribution and partition of As may exhibit a large variation at different depths in sediments, which is closely

related to iron oxyhydroxides redox cycling, the availability of manganese oxides and organic matter, the thickness of the oxic layer and its co-precipitation with sulfides and iron sulfides (Mucci et al., 2000; Rubinos et al., 2003; Bennett et al., 2012; Aziz et al., 2016). Recently, Sun et al. (2016a, 2016b) and Wang et al. (2016a, 2016b) investigated the distribution of labile As in sediments of two lakes (Lakes Taihu and Hongze, China) at a vertical resolution of 2 mm using a passive sampling technique called diffusive gradients in thin films (DGT). The results showed an initially increasing phase of the DGT-labile As with depth in shallower layers, usually followed by a decreasing phase in deeper layers. The maximal concentration of DGT-labile As was one to two orders of magnitude greater than that at the sediment-water interface (SWI). Yao et al. (2016) further found that the heterogeneous distribution of DGT-labile As could be caused by macrozoobenthos bioturbation and hydrodynamics due to the changing oxic-anoxic conditions. These studies showed a great necessity in the study of As biogeochemical processes by taking the heterogeneous distribution characteristics into consideration (Gorny et al., 2015). It needs to be noted that, until now, the dynamic change of As in sediments under the competition of P has remained unclear using the previous batch experiments (Gao et al., 2011; Rubinos et al., 2011). A further direct observation of the competition effect in sediments at a millimeter scale has not been available with regard to the heterogeneous distribution of As, which should be key to the understanding of the mechanism of this effect.

In this study, an incubation experiment was performed to investigate the effects of elevated P content in water on the mobility of As in sediments on a millimeter scale. High-resolution dialysis (Peeper) (Ding et al., 2010) and DGT (Sun et al., 2015) techniques were employed to simultaneously measure dissolved P, Fe and As in pore water and labile P, Fe and As mainly released from sediment solids, respectively. The competitive effect of P was assessed based on its concentration changes, together with the changes of As speciation in sediments. The DGT induced fluxes in sediments (DIFS) model was further used to characterize the kinetics of As release in sediments.

2. Materials and methods

2.1. Preparation of Peeper and DGT probes

The Peeper was used for measurements of dissolved Fe, P and As in pore water of sediment. The construction of the Peeper device has been reported by Ding et al., 2010. It has a vertical resolution of 4 mm for sampling. Prior to use, the Peeper device was soaked in 10% (v/v) nitric acid overnight and rinsed thoroughly using deionized water. The chambers of the device were filled with deionized water, followed by coverage with a cellulose nitrate membrane (Whatman, $0.45 \mu\text{m}$ pore size) and a window frame at each of the two exposure sides. The loaded Peeper was stored in deionized water prior to use.

The ZrO-Chelex DGT was provided by Easysensor Ltd. (www.easysensor.net). It is a flat type DGT device loaded with a mixed ZrO-Chelex binding gel for simultaneous measurements of labile Fe, P and As in sediments (Sun et al., 2015). Its measurement can reflect pore water concentrations of Fe, P and As and their dynamic resupply from sediment solids taking the heterogeneous nature of the sediment into consideration (Stockdale et al., 2009; Zhang et al., 2014). The ZrO-Chelex binding gel was prepared according to the procedure reported by Xu et al. (2013). The agarose diffusive gel was prepared according to Wang et al., 2016a,b. A new flat-type holder was used for assembling the DGT probe, which has a low disturbance on the sediment during the probe insertion (Ding et al., 2016). For assembling the DGT probe, the Durapore PVDF

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