



## Arsenic speciation in aquifer sediment under varying groundwater regime and redox conditions at Jiangnan Plain of Central China



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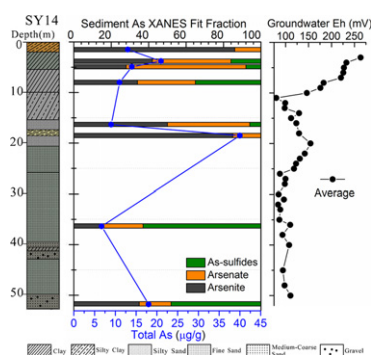
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### HIGHLIGHTS

- Arsenic speciation was determined by linear combination fits of As K-edge XANES.
- Sediment As speciation mainly controlled by redox conditions in the groundwater system.
- Groundwater regime and redox conditions greatly affected As content and speciation in the sediments.
- Sediment As speciation and reactivity controlled groundwater As concentration.

### GRAPHICAL ABSTRACT



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### ABSTRACT

At Jiangnan Plain of central Yangtze basins where the health of >73,000 people has been affected by long term intake of high arsenic groundwater, over 100 sediment samples from four boreholes at the field monitoring sites were collected and analyzed to delineate the distribution and speciation of As in the shallow aquifer sediment. Results showed that sediment As concentration is generally dependent on the lithological conditions, with the higher As concentration present in fine particle sediment, especially in the silty sand layers underlying clay or silty clay layers. High As concentration in the sediment mainly occurred in three different depth ranges: <5 m, 15–35 m, and >35 m. Both the groundwater regime and redox conditions played important roles in controlling sediment As speciation. Arsenate (86%) was the dominated As species in the near surface sediment. As the redox turned to be reducing, arsenite (64%) became the dominant species in the underlying clay and silty clay layers. But in the silty sand aquifer near the boundary of unconfined aquifer and confined aquifer, arsenate (85%) became the dominant species again as results of redox potential elevation. In the deep medium to coarse sand aquifers (>35 m deep), As-sulfides (49%–63%) were the main species of As. The speciation and reactivity of sediment As strongly controlled the spatial distribution of groundwater As concentration, while seasonal variation in groundwater As concentration and speciation affected the content and speciation of sediment As.

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## 1. Introduction

Geogenic arsenic (As) pollution of drinking water has been reported in >70 countries, posing a serious health hazard to an estimated 150 million people world-wide (Brammer and Ravenscroft, 2009), and 19.6 million people in China (Rodriguez-Lado et al., 2013). Long time exposure of As may cause various diseases such as melanosis, keratosis, non-petting oedema, gangrene, leucomelanosis, respiration system problems and an increased cancer risk for skin, bladder, and lungs (Chen and Ahsan, 2004; Chowdhury et al., 2000; Kapaj et al., 2006). As also causes cardiovascular disease and inhibits children mental development (Chen et al., 1996; Wasserman et al., 2004).

Sediment As content and speciation are the two critical factors controlling As partitioning and transport. In oxidized aquifer sediment, As(V) adsorbs more strongly than As(III), and As(III) could be oxidized to As(V), leading to strong As retention (Stollenwerk et al., 2007). Under reducing conditions, As can be released from colloids or Fe/Mn oxide surfaces through desorption, reductive dissolution of As-containing Fe (III) minerals, and As(V) reduction to more labile As(III), which subsequently enhances As mobility and toxicity in the aquifers (Bhattacharya et al., 1997, 2002; Donselaar et al., 2017; Fendorf et al., 2010; Guo et al., 2008; Nickson et al., 2005; Paul et al., 2015; von Brömssen et al., 2007; Zheng et al., 2004). However, As can be removed from the aqueous phase via precipitation or incorporation of As-Fe sulfides under reducing conditions (Bostick et al., 2004; Lowers et al., 2007; O'Day et al., 2004). Sorption of both As(V) and As(III) is strong on Fe(III) oxides under circumneutral pH conditions (Dixit and Hering, 2003), small goethite particles can enhance As sorption and/or coprecipitation during As fixation (Xie et al., 2016). Incorporation of As in carbonates can effectively mobilize As from sediment into groundwater (Anwar et al., 2004; Kim et al., 2000; Saunders et al., 2005; Sengupta et al., 2004). Additionally, As(V) can substitute for  $\text{SiO}_4^{4-}$  within silicate minerals (Seddique et al., 2008), and As(V) is retained more appreciably than As(III) on silicate minerals, such as chlorite and halloysite (Lin and Puls, 2000). These previous results indicate that sediment As speciation directly controls As concentration, mobility and toxicity in groundwater. Characterizing the distribution and speciation of As in the sediment is therefore critical for understanding the speciation and spatial variation of groundwater As.

Since the first reports of waterborne As poisoning in Shahu Village within the Jiangnan Plain in 2005, studies by Deng et al., 2014, Y. Gan et al., 2014; Y.Q. Gan et al., 2014, Duan et al., 2015, Li et al., 2015, and Schaefer et al., 2016 have made efforts to understand the processes and factors leading to high concentrations of As in groundwater. The elevated As concentration in groundwater has affected the health of >73,000 people, including 20,000 children (Li et al., 2010; Wang and Zhao, 2007). Our previous work was mostly focused on the effects of groundwater hydrogeochemistry on As distribution (Y. Gan et al., 2014; Duan et al., 2014) and dynamic changes in groundwater As concentration (Deng et al., 2014; Duan et al., 2015; Y.Q. Gan et al., 2014; Schaefer et al., 2016). Although a great deal of effort has been made in most high arsenic groundwater regions to investigate the mineralogy and geochemistry of the aquifers sediment, and sediment-associated As that are expected to have great impacts on As enrichment in groundwater (Bhattacharya et al., 2001; Hasan et al., 2009; Hossain et al., 2014; Mukherjee et al., 2009; Neidhardt et al., 2013; von Brömssen et al., 2008), litter work has been done in the Jiangnan Plain.

The results of previous work indicated that the sediment samples from Shahu Village contained As concentration ranging from 1 to 107 mg/kg (Duan et al., 2014; Li et al., 2015), with a median of 12.8 mg/kg (Li et al., 2015). Higher As concentration was observed in the silt and clay samples, with the highest concentration near 20 m depth. All sediment samples had high concentrations of Fe and Mn, with  $\text{Fe}_2\text{O}_3$  contents ranging from 4% to 9% (Y. Gan et al., 2014). Li et al. (2015) also reported that 10%–70% (with a median value of 38%) As in the sediment could be extracted by ammonium oxalate, implying

that a large amount As in the sediment was bound with amorphous Fe oxides. The main mineralogical compositions in the sediment were clay minerals (montmorillonite, chlorite, illinite and kaolinite) and quartz (Y. Gan et al., 2014). And the main source of As in the sediment at Jiangnan Plain was thought to be the As-bearing minerals (As-bearing Fe oxides, and As-bearing sulfides) deposited during fluvial-lacustrine deposition (Li et al., 2015).

The objectives of this research are: (1) to understand the hydrostratigraphy and the relationship with the sediment types; (2) to determine As concentration and speciation in sediment as a function of depth in the aquifer; and (3) to correlate sediment-associated As with groundwater As to provide insights into the factors controlling arsenic concentration in groundwater at Jiangnan Plain.

## 2. Materials and methods

### 2.1. Site describing

Jiangnan Plain is an alluvial plain formed by the Yangtze and Han rivers located in the Middle Reaches of the Yangtze River that includes the central and southern regions of Hubei Province (Fig. 1). It has a subtropical monsoonal climate, the average annual precipitation increases from 800 mm in the northwest to 1500 mm or more in the southeast, 30–50% of which occurs in summer. Jiangnan Plain is a semi-closed Quaternary basin with a higher elevation in the north and a lower elevation in the south.

The field monitoring site for this study was constructed in Shahu Village, the interior of the Jiangnan Plain (Fig. 1). The site is surrounded by four rivers (Tongshun River, Dongjing River, Kuige River, and Lüfeng River), and covered by other abundant surface water bodies such as ponds, irrigation channel, and wetlands. Surface water levels at the field site fluctuate 5.6–7.6 m annually and are closely tied to monsoon rains. Well water levels follow a trend similar to surface water, but the magnitude of the fluctuations is approximately 0.7–2.4 m and the timing of falling and rising well water levels lags surface water changes (Duan et al., 2015; Schaefer et al., 2016). Strong surface water-groundwater interactions were observed here. During the wet summer monsoon when surface water levels are higher than groundwater levels, a groundwater recharge flow gradient develops; during drier, winter months, surface water levels drop below groundwater levels, and groundwater flow reverses and moves toward surface water. Further details of the field area are provided in previous publications (Duan et al., 2015; Deng et al., 2015; Schaefer et al., 2016).

### 2.2. Sampling

Groundwater samples, 729 in total, were collected in Xiantao, Honghu, Qianjiang, and Jianli in the Jiangnan Plain from 2011 to 2014. Temperature, conductivity, pH and oxidation-reduction potential (ORP) were measured in the field using a HACH HQ40D multimeter. ORP was converted to Eh depending on the water temperature. Samples were filtered and acidified with concentrated HCl for analysis of total dissolved As concentration.

Sediment samples were collected during well installation in the Shahu field site from boreholes SY15 (N 30°09'23.73", E 113°40'42.59"), SY03 (N 30°09'23.51", E 113°40'39.21"), SY14 (N 30°09'18.62", E 113°40'21.03"), and SY07 (N 30°09'19.66", E 113°39'58.76") (Fig. 1). Sediment samples from SY03 and SY07 were collected in November 2011, samples from SY14 and SY15 were collected in November 2014. A Geoprobe DT21 rig was used to get continuous *in-situ* sediment samples from depths < 20 m. The sediment (18 cm long) in sampling tube were cut and sealed for lab analysis. A rotary drill rig and split-tube sampler with an internal stainless steel tube (17.5 cm length, 3.8 cm diameter) was used to obtain sediment samples > 20 m deep. After sample collection, both ends of the sampling tubes were covered with polytetrafluoroethylene caps and sealed with paraffin wax,

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