



Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: [www.elsevier.com/locate/scitotenv](http://www.elsevier.com/locate/scitotenv)

## Arsenic behavior in different biogeochemical zonation approximately along the groundwater flow path in Datong Basin, northern China

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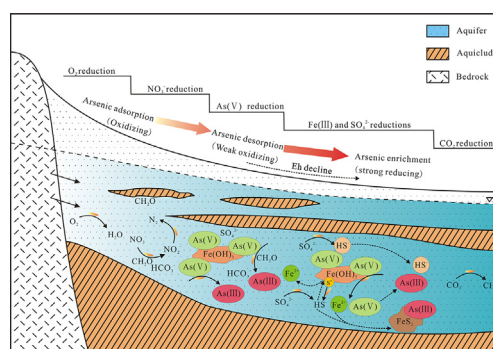
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### HIGHLIGHTS

- Bacterial community structures are distinctly different along groundwater flow path.
- Bacterial activities play key roles in groundwater redox transformation.
- $\text{NO}_3^-$  reduction process inhibits arsenic release, while  $\text{SO}_4^{2-}$  process doesn't.
- Reductive desorption of As(V) is an important process in weakly alkaline groundwater.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 25 October 2016

Received in revised form 30 December 2016

Accepted 5 January 2017

Available online xxxx

Editor: D. Barcelo

#### Keywords:

Arsenic

Biogeochemistry

Hydrogeochemistry

Redox environment

### ABSTRACT

Studies have shown that arsenic is desorbed/released into groundwater as a result of bacterial reduction of As(V) and Fe(III). However, bacterial activities like sulfate reduction process can also reduce the content of arsenic in groundwater. In this study, we examined the effects of different biogeochemical processes (e.g.  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  reduction) on arsenic, by investigating the chemical characteristics and bacterial community structure of groundwater in the Datong Basin, northern China. Along the groundwater flow path, arsenic concentration increased from  $<1$  to  $947.6 \mu\text{g/L}$  with dominant bacteria change from aerobic (*Fluviicola*, *Rhodococcus*) to denitrifying bacteria (*Thauera*, *Gallionella*), and then to sulfate reducing bacteria (*Desulfosporosinus*). According to the groundwater redox sensitive indicators (Eh,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}/\text{Cl}^-$  and  $\text{Fe}^{2+}$ ) concentrations (or ratios), the sampling points were approximately divided into three zones (I, I' and II). Variation in features of these indicators suggested that the groundwater evolved from a weakly oxidizing environment (Zone I, Eh average 93.3 mV, respectively) to strong reducing environment (Zone II, Eh average  $-101.8$  mV). In Zone I, bacteria mainly consuming  $\text{O}_2$  or  $\text{NO}_3^-$  were found which inhibits Fe(III) and As(V) reduction reaction, resulting in a low As zone ( $<1$  to  $3.3 \mu\text{g/L}$ ). However, in Zone II, where  $\text{O}_2$  and  $\text{NO}_3^-$  have been depleted,  $\text{SO}_4^{2-}$  reduction appears to be the dominant process, and the Fe(III) and As(V) reduction processes are also occurring and hence, enrichment of As in the groundwater ( $2.8$  to  $947.6 \mu\text{g/L}$ , average  $285.6 \mu\text{g/L}$ ). Besides, bacterial Fe(III) reduction process was retarded due to the weakly alkaline conditions (pH 7.60–8.11, average 7.83), but abiotic Fe(III) reduction by  $\text{HS}^-$  may be continued. Therefore, we conclude that the Fe(III) and As(V) reduction processes contributed to arsenic enrichment in the groundwater, and the reductive desorption of arsenate is the main occurring process especially in the weakly alkaline

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environment. Moreover,  $\text{NO}_3^-$  reduction process can significantly restrain the release of arsenic, but the process of  $\text{SO}_4^{2-}$  reduction is insignificant for arsenic concentration decline in natural groundwater.

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## 1. Introduction

Natural occurrence of arsenic enrichment in groundwater has been documented worldwide, including Bangladesh, India, America, Hungary, Romania and China (Nickson et al., 2000; Nordstrom, 2002; Smedley and Kinniburgh, 2002; Berg et al., 2007). This geogenic release of arsenic into groundwater has resulted in sustained health related issues (including skin and lung cancer), and presently poses a threat to the livelihood of tens of millions across the globe (Chakraborti et al., 2016). Therefore, in the past 20 years, numerous studies have been dedicated to probing the cause of arsenic release into groundwater and outcomes of these studies furnish three main mechanisms which are believed to be the culprit of this problem: First, the oxidation of arsenic-bearing pyrite (Chowdhury et al., 1999); second, desorption by competitive anions (such as  $\text{PO}_4^{3-}$ ,  $\text{HCO}_3^-$ ,  $\text{OH}^-$ ) (Acharyya et al., 1999; Appelo et al., 2002; Jain et al., 1999); and third, is bacteria activity (Nickson et al., 2000; Zobrist et al., 2000; Langner and Inskeep, 2000). However, increasing research in this field have ushered in a new understanding that bacterially mediated dissimilatory reduction of arsenic-bearing Fe (hydrogen) oxides minerals and reductive desorption of arsenate (energy-conserving respiratory) is the major cause of arsenic enrichment in shallow aquifers (Cummings et al., 1999; Oremland and Stolz, 2003; Islam et al., 2004; Oremland and Stolz, 2005; Campbell et al., 2006; Kudo et al., 2013; Yadav et al., 2015; Paul et al., 2015; Das et al., 2016).

Although copious evidences have shown that bacteria play an important role in the release of the arsenic, some others indicated that bacterial activities can also reduce the content of the arsenic in groundwater. The sulfide produced during bacterial  $\text{SO}_4^{2-}$  reduction can form minerals that remove arsenic from solution (Rittle et al., 1995). Geochemical evidence suggests that the activity of  $\text{SO}_4^{2-}$ -reducing bacteria play an important role for arsenic accumulation to hazardous levels (Kirk et al., 2004). In addition, the Fe (hydrogen) oxides minerals produced by bacterial  $\text{NO}_3^-$  reduction ( $\text{Fe}^{2+}$  oxidation) can also remove arsenic. In a field test, Harvey et al. (2002) observed arsenic content decreased coinciding with the injection of nitrate solution into the aquifer. They concluded that this phenomenon was probably as a result of arsenic adsorption onto iron oxyhydroxides which is precipitated by bacterial process. Moreover, laboratory experiments have demonstrated that bacterial mediated sulfate-reducing and nitrate-reducing processes can both remove arsenic from solution (Keimowitz et al., 2007; Sun et al., 2009; Omeregie et al., 2013). The works described above show that bacterial activities can promote arsenic release/desorption (Fe(III) and As(V) reduction) into groundwater and also can remove arsenic ( $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  reduction) from groundwater. It is therefore worthwhile to study the different biogeochemical processes and their effects on arsenic geochemical behavior in natural aquifers.

Typically, bacteria consume organic matter to obtain energy with oxygen as an electron acceptor in aerobic environment. When oxygen is depleted, other oxygen-bearing species becomes an alternative for consumption. Chapelle (2000) indicated that bacteria in groundwater consumed DO (dissolved oxygen), nitrate, Fe(III), sulfate and  $\text{CO}_2$  in turn along the direction of groundwater flow, evidencing different biogeochemical trend with the groundwater environment evolving from oxidative to reductive conditions. The adsorption, release and co-precipitation of arsenic in groundwater are closely related to the content of DO,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , Fe(III)/ $\text{Fe}^{2+}$  and DOC (dissolved organic carbon) (Fendorf et al., 2010; Sharif et al., 2008; Harvey et al., 2006). But the content of these components changes with the flow, owing to changes in hydrological or biogeochemical process. Many studies have investigated

the content of these components, the isotopic composition and the change of the redox environment along the direction of groundwater flow to explain the release mechanism of arsenic under the activity of bacteria (Jia et al., 2014; Guo et al., 2011; Xie et al., 2009; Benner et al., 2008; Stute et al., 2007; Postma et al., 2007). Nevertheless, only few studies have investigated the bacterial community structure along the groundwater flow path and examined the effects of different biogeochemical processes on arsenic.

In this study, the groundwater and bacteria samples were collected and isolated respectively along the direction of groundwater flow at a known high-arsenic area in the Datong Basin and evaluated the following objectives: (1) the bacteria community structure information, (2) observed the variable characteristics of the arsenic and other redox sensitive indicators contents, (3) unraveled the cause of redox environment evolution and revealed arsenic behavior under different biogeochemical conditions.

## 2. Study area

Geologically, Datong Basin is a Cenozoic Basin located in the semi-arid region of northern China (Wang and Shpeyzer, 2000). This Basin extends from southwest to northeast and is surrounded by the Hengshan Mountains, the Guancen Mountains and the Hongshou Mountains (Xie et al., 2013a). The piedmont is the main bedrock, while the central basin consists of Quaternary unconsolidated sediments with thickness up to approximately 400 m. According to the burial depths of aquifers, the Quaternary groundwater systems can be divided into three groups: upper (5–60 m), middle (60–160 m) and lower aquifers (>160 m) (Wang et al., 2009). The grain size of the sediment generally decreases from margin to the center of the basin. The central basin is lacustrine and alluvial-lacustrine sandy loam, silt and silty clay with high content of organic matter. The organic matter content reaches as high as 1% in the aquifer sediments (Guo et al., 2003), while the TOC (total organic carbon) concentration reaches up to 17.5 mg/L in groundwater (Luo et al., 2012).

The groundwater is mainly recharged by vertically infiltrating meteoric water and laterally penetrating fracture water from the bedrock in the domain of the mountains (Wang et al., 2009). Discharge occurs mainly via evapotranspiration and artificial abstraction. The Rivers are ephemeral and emerge in July to August (monsoon), while precipitation is the main recharge. There are two general flow regimes at Datong Basin: (1) flow from margins mountain front areas to the central of the basin, and (2) flow inside the basin along the direction of the river from southwest to northeast (Xie et al., 2009). The velocity of the groundwater movement is relatively faster in mountain front areas than the central portion of the basin due to the aquifers characteristics, which are mostly gravel and coarse sand with high hydraulic gradient in mountain front areas (Wang et al., 2009). Compared to the mountain fronts, the terrain is flat towards the basin center, while the aquifers are mainly lacustrine and alluvial-lacustrine fine sand and silty sand with low hydraulic gradient. The groundwater movement is relatively slow and the velocity ranges from 0.20 m to 0.58 m per day ( $K = 20\text{--}58$  m per day,  $J < 1/100$ ) (Xie et al., 2009). The depth of water table is commonly <2 to 5 m in the central portion of the basin, with an artesian well (SHY-02) observed within the survey area.

## 3. Sampling and analytical methods

A total of twenty-two water samples were collected along the groundwater flow path in August 2014 at the Datong basin. According

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