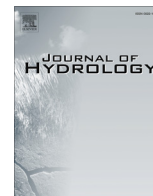




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Coupled iron, sulfur and carbon isotope evidences for arsenic enrichment in groundwater



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SUMMARY

It is generally accepted that microbial processes play a key role in the mobilization and enrichment of arsenic (As) in groundwater. However, the detailed mechanism of the metabolic processes remain poorly understood. We apply isotopic measurements of iron ($\delta^{56}\text{Fe}$ vs. IRMM-14), sulfur ($\delta^{34}\text{S}_{\text{SO}_4}$ vs. V-CDT) and carbon ($\delta^{13}\text{C}_{\text{DIC}}$ vs. V-PDB) to an experimental field plot in the Datong Basin, northern China. An array of monitoring wells was installed in a $\approx 1700\text{-m}^2$ plot in which high concentrations of As, ranging from 4.76 to 469.5 $\mu\text{g/L}$, were detected in the groundwater. The measured range of $\delta^{34}\text{S}_{\text{SO}_4}$ values from 10.0‰ to 24.7‰ indicates the prevalence of microbial sulfate reduction within aquifers. The range of $\delta^{56}\text{Fe}$ values measured in the groundwater suggests microbial Fe(III) reduction and the occurrence of isotopic exchange between Fe(II)aq and FeS precipitation. The low $\delta^{13}\text{C}_{\text{DIC}}$ values (up to -33.6‰) measured in groundwater are evidences for the microbial oxidation of organic matter, which is interpreted as the light carbon pool within the aquifer sediments. The high As ($\text{As} > 50 \mu\text{g/L}$) groundwater, which has higher $\delta^{34}\text{S}_{\text{SO}_4}$ and $\delta^{56}\text{Fe}$ values and lower $\delta^{13}\text{C}$ values, indicates the following: (1) microbial reduction of sulfate causes the mobilization of As through HS^- abiotic reduction of Fe(III) minerals and/or formation of As-sulfur components; and (2) direct microbial reduction of Fe(III) oxides, hydroxides and oxyhydroxides cannot increase As concentrations to greater than 50 $\mu\text{g/L}$. Re-oxidation of Fe-sulfide explains how sample C1-2 can have a high As concentration and low $\delta^{34}\text{S}_{\text{SO}_4}$ and high $\delta^{56}\text{Fe}$ values. The results provide new insight into the mechanism of As enrichment in groundwater.

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1. Introduction

Arsenic (As) contamination of groundwater in sedimentary aquifers is a major global public health crisis. Natural occurrences of high As groundwater have been reported in several geologically young aquifers around the world, including the Ganges Plain (India and Bangladesh) (Harvey et al., 2002; Islam et al., 2004; Nickson et al., 2000), the Red River Delta (Vietnam) (Berg et al., 2007, 2008; Postma et al., 2007), the Mekong River Basin (Cambodia) (Buschmann et al., 2008; Papacostas et al., 2008; Thi et al., 2010) and the Hetao Basin in China (Guo et al., 2008a,b). Similar conditions are also present in the Datong Basin of Shanxi Province, north China (Wang et al., 2009; Xie et al., 2009). Arsenic concentrations as high as 1820 $\mu\text{g/L}$ have been detected in groundwater of the

Datong Basin (Xie et al., 2008), which greatly exceeds the maximum limited value (10 $\mu\text{g/L}$) for drinking water recommended by the WHO (WHO, 2004).

Aquifers with natural As contamination are characterized by As concentrations that vary significantly over short lateral and vertical distances (Kirk et al., 2004). Studies of As-contaminated aquifers around the world demonstrate that As concentrations in groundwater are controlled by complex sets of conditions and biogeochemical processes (Appelo et al., 2002; Bose and Sharma, 2002; Islam et al., 2004; Kirk et al., 2004; Nickson et al., 2000; Oremland and Stolz, 2003; Polizzotto et al., 2008). Arsenic that is sorbed and sequestered on Fe oxides/hydroxides is one of the most common As reservoirs in some sedimentary basins. The reduction of As(V) to As(III) results in desorption or the formation of weakly sorbing complexes. Changes in pH and redox conditions may induce the dissolution of Fe-oxides, hydroxides and oxyhydroxides and/or the release of sorbed As from the mineral surfaces (Nickson et al., 1998, 2000; Swartz et al., 2004). And the formation of

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As-sulfur compounds in high HS^- concentration water promote the mobilization of As (Planer-Friedrich et al., 2007; Stauder et al., 2005; Bostick et al., 2005). In addition, biotic processes can play important roles by either directly mediating redox reactions or changing the redox conditions in the aquifer (Oremland and Stolz, 2003). More recently, Polizzotto et al. (2008) proposed that surface hydrological processes and groundwater flow can significantly promote the enrichment of As in groundwater. Accordingly, the mechanism of As release may vary with location depending on the hydrogeological conditions and (bio)geochemical processes. This variation can be accounted for by the distribution of As concentrations in natural As-contaminated aquifers, which is crucial to understanding the generation of high As groundwater. However, this explanation is complicated by heterogeneities in sediment lithology and variations in the hydrogeological conditions and (bio)geochemical processes within the aquifers. Therefore, the complex (bio)geochemical processes should be investigated by analyzing the different hydrogeochemical constituents (including the isotopic compositions) of groundwater in a high As aquifer system in a relatively homogenous hydrogeological setting.

Hydrogeochemical studies of a field experimental plot (20 m × 85 m) in the Datong Basin of Shanxi Province, China may provide a clear understanding of the complex (bio)geochemical processes that cause As mobilization. The experimental plot is located in a groundwater discharge zone in the center of the basin and adjacent to a river. The groundwater at this site moves from south to north toward the river (Fig. 1). Because the area is far from mountains, no significant mixing of different recharge waters is expected at the study site. Studies of sediment cores from the experimental plot indicate that the sediment consist of homogeneous fine sands and silts. Three sand layers are present at depths

less than 30 m below the surface. Arsenic is associated with Fe in the sediments at this site, and the Fe in the sediments is mainly present as crystalline Fe(III) oxides/hydroxides (Xie et al., 2013a). Microbial Fe(III) and SO_4^{2-} reduction is thought to be the controlling process that causes the mobilization of As within the aquifers (Xie et al., 2013a). This interpretation is supported by the low ORP (oxidation and reduction potential) values and the high concentrations of Fe(II) and HS^- in the groundwater (Xie et al., 2013b). However, the detailed metabolic mechanisms of Fe and S and the effects on As mobilization are not clear. The relative homogeneity of the sediments and the simple hydrogeological conditions at the site provide excellent conditions for investigating the (bio)geochemical effects on As mobility.

The (bio)geochemical cycles of As are closely linked with S and Fe (Fisher et al., 2008; Oremland and Stolz, 2003). Sulfate-reducing bacteria use sulfate (SO_4^{2-}) as a terminal electron acceptor to oxidize organic matter and produce sulfide (HS^-) (Postgate, 1984). Sulfur isotopes of sulfate ($\delta^{34}\text{S}_{\text{SO}_4}$) are fractionated during SO_4^{2-} reduction, and the residual SO_4^{2-} is enriched in the heavier isotope (Böttcher and Thamdrup, 2001; Canfield, 2001; Detmers et al., 2001; Robertson and Schiff, 1994). Similarly, Fe(III)-reducing bacteria oxidize organic matter by using ferric oxide minerals as terminal electron acceptors (Canfield et al., 1993; Lovley, 1997; Raiswell and Canfield, 1998) to produce dissolved Fe(II). In natural environments, an Fe isotopic fingerprint appears to be associated with microbial Fe reduction in the form of low $\delta^{56}\text{Fe}$ values for aqueous Fe(II) (Beard et al., 2003; Percak-Dennett et al., 2011). The geochemical cycles of Fe and S are strongly linked, so studies of Fe and S isotope systems should provide a unique view of the metabolic processes of Fe and S. Sulfur and Fe isotopes are therefore useful tools for tracing biogeochemical processes in the

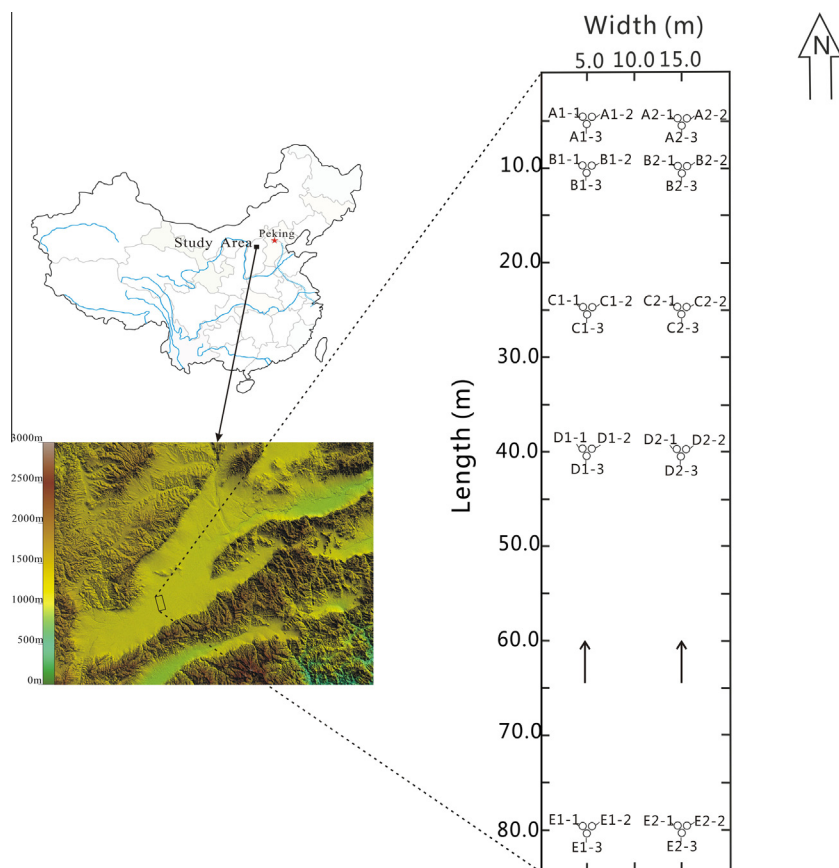


Fig. 1. Location of the experimental plot. The monitoring wells are indicated. The direction of groundwater flow from south to north is noted by the arrow on the map.

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