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Impact of sedimentary provenance and weathering on arsenic distribution in aquifers of the Datong basin, China: Constraints from elemental geochemistry

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SUMMARY

Arsenic (As)-contaminated aquifer sediments from Datong basin, China have been analyzed to infer the provenance and depositional environment related to As distribution in the aquifer sediments. The As content in the sediments ranged from 2.45 to 27.38 mg/kg with an average value of 9.54 mg/kg, which is comparable to the average value in modern unconsolidated sediments. However, minor variation in As concentration with depth has been observed in the core. There was a significant correlation between Fe, and Al and As, which was attributed to the adsorption or co-precipitation of As onto/with Fe oxides/hydroxides and/or Fe-coated clay minerals. Post-Archean Australian Shale (PAAS)-normalized REEs patterns of sediment samples along the borehole were constant, and the sediments had a notably restricted range of La_N/Yb_N ratios from 0.7 to 1.0. These results suggested that the provenance of the Datong basin remained similar throughout the whole depositional period. The analysis of major geochemical compositions confirmed that all core sediments were from the same sedimentary source and experienced significant sedimentary recycling. The co-variation of As, V/Al, Ni/Al and chemical index of alteration (CIA) values in the sediments along the borehole suggested that As distribution in the sediments was primarily controlled by weathering processes. The calculated CIA values of the sediments along the borehole indicate that a relative strong chemical weathering occurred during the deposition of sediments at depths of ~35 to 88 m, which was corresponding to the depth at which high As groundwater was observed at the site. Strong chemical weathering favored the deposition of Fe-bearing minerals including poorly crystalline and crystalline Fe oxide mineral phases and concomitant co-precipitation of As with these minerals in the sediments. Subsequent reductive dissolution of As-bearing poorly crystalline and crystalline Fe oxides would result in the enrichment of As in groundwater. In general, the chemical weathering during the deposition of the sediments governed the co-accumulation of Fe oxides and As in the aquifer sediments. And then, the reductive dissolution of Fe oxides/hydroxides is the mechanism of As enrichment in the groundwater in the Datong basin.

1. Introduction

Arsenic (As)-contaminated groundwater in sedimentary aquifers has been extensively reported worldwide (Ahmed et al., 2004; Brammer and Ravenscroft, 2009; Bhattacharya et al., 2004, 2007; Chatterjee et al., 2010; Erban et al., 2013; Fendorf et al., 2010; Mukherjee-Goswami et al., 2012; Polya et al., 2008; Rodriguez-Lado et al., 2013; Shukla et al., 2010; Winkel et al., 2008) and has caused serious health problems in millions of people in the last few decades (Charlet and Polya, 2006; Kar-Purkayastha, 2012; Kapaj et al., 2006; Rahman et al., 2005; Roychowdhury, 2010; Yu et al., 2007). In the Datong basin, groundwater is acritical drinking and irrigation water source. Unfortunately, the groundwater at the basin contains high concentrations of soluble As, which was up to 1820 μg/L, (Xie et al., 2008), threatening the health of tens of thousands of local residents that depend on it (Zhao et al., 2005). To eliminate or mitigate the impacts of this contamination, it is imperative to understand the distribution and generation of As-contaminated groundwater in this area.

The mobilization of As in sedimentary aquifers is mainly influenced by local geology, hydrogeology, geochemistry of sediments and water, as well as anthropogenic activities, such as irrigation

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practices (Zheng et al., 2004; Wang et al., 2009; Xie et al., 2012a, 2012b Bhattacharya et al., 2004, 2007). In Datong basin, the occurrence of high levels of As in the groundwater is attributed to the prevalence of a strongly reducing subsurface environment (Anawar et al., 2002; Akai et al., 2004; Xie et al., 2009a). Recent studies suggested that reductive dissolution of Fe-oxides/hydroxides is the key mechanism that enriches As in groundwater in Datong sedimentary aquifers (Xie et al., 2013a, 2013b). The organic matter in the aquifer sediments may also have played an important role in As mobilization (Xie et al., 2012a).

The geogenic occurrence of high As concentrations in groundwater is generally believed to be the result of water–sediment interactions (Haque et al., 2008; Liu et al., 2009; Smedley and Kinniburgh, 2002). The As in sedimentary aquifers may result from chemical and mechanical weathering of As-bearing rocks (Saunders et al., 2005; Xie et al., 2011; Zheng, 2007), and As accumulation and distribution in aquifer sediments is often determined by provenance and weathering intensity. In the Datong basin, the aquifer sediments were largely derived from chemical weathering and subsequent erosion of bedrock around the basin in the Pliocene to Pleistocene and Holocene (Li and Zhu, 2000). A few studies that were performed on bulk sediments indicated that the presence of high As contents in sediments is not associated with primary As-bearing minerals, rather mostly appears to be from secondary phases including poorly crystalline and crystalline Fe oxides (Xie et al., 2013a, 2008), which are closely related to the depositional environments.

Although considerable studies have been conducted to understand the occurrence of high As groundwater in this area, the importance of chemical weathering intensity and provenance on As enrichment in groundwater is still not well understood. Recent studies have suggested that sedimentary geochemical elemental records may contain additional information that may provide important insights into sediment chemical weathering intensity and provenance (Cullers, 2002; Qiao et al., 2011; Wei et al., 2004). In this paper, we analyzed the geochemistry of major and trace elements in the bulk sediments collected from a known As-contaminated site in the Datong basin to derive the relationships between high concentrations of dissolved As in aquifers and sediment weathering processes during sediment deposition.

2. Geological and hydrogeological setting

The Datong basin is located in the northern part of China and bounded by Hengshan Mountains, Guancen Mountains and Hongshou Mountains in the southeast, west and northwest of the basin, respectively (Fig. 1). The Hengshan Mountains mainly consist of the Neoarchaean Hengshan Complex, which consists of migmatitic, tonalitic, granodioritic and granitic gneiss, as well as mafic lenses, pegmatites and granites (Li and Qian, 1994). There are Lower Paleozoic (Cambrian and Ordovician) limestone outcrops in the Guancen Mountains. The Hongshou Mountains are primarily Permian to Cretaceous clastic rocks including sandstone, shale as well as coal. Datong volcanic field Late Pleistocene basalt occurs in the northern part of the basin. In the western margin of the basin, Quaternary aquifer sediments lie over Cambrian and Ordovician limestone and younger sedimentary rocks. The Quaternary sediments are underlain by fractured Archaean metamorphic rocks in the eastern margin of the basin.

The basin is composed of Pliocene to Pleistocene and Holocene unconsolidated sediments with thickness varying from 1500 m to 3500 m (Li and Zhu, 2000). Analysis of mollusks fossils and microfossils (Ostracoda fossils) assemblages revealed four major stratigraphic units (Table 1) in the Quaternary sediments (Q_1–Q_4) (Liu et al., 1986): (1) Early Pliocene stratigraphic unit, which was produced in stable and weak hydrodynamic conditions of the palaeo-lake expansion, and consisted of a sedimentary sequence ofclay to silty clay with grayish to gray-green color. The thickness of this sedimentary sequence is generally larger than 200 m; (2) Early to Middle Pliocene stratigraphic unit: This sedimentary sequence has thicknesses ranging from 20 to 30 m and is composed of grayish-yellow silt and silty clay. The uppermost part of the sequence is fine to medium sand. The deposit environment is typical, deep to shallow lake, which is characterized by the stable

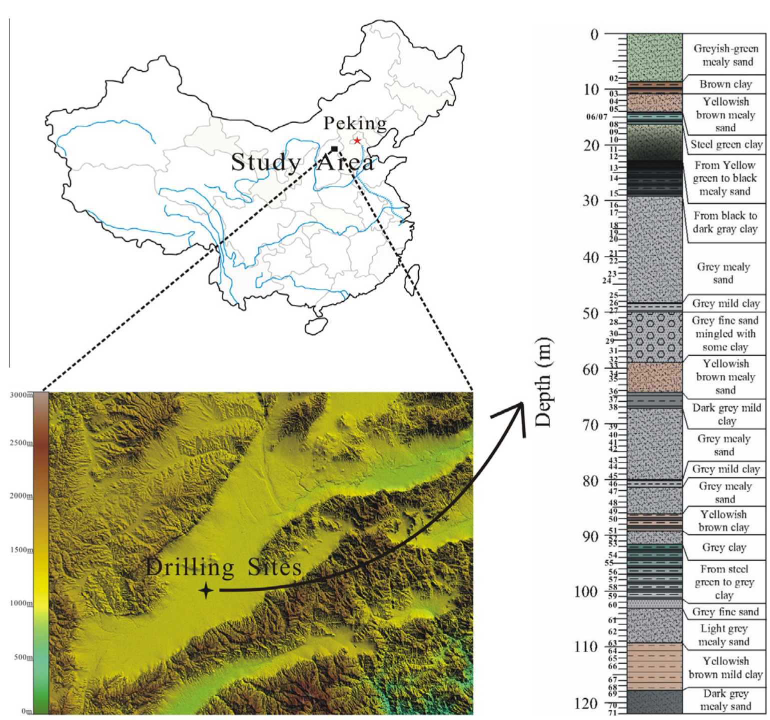

Fig. 1. Location of the borehole used for sediment samples and the lithological profiles of the borehole.