Sedimentological controls on the formation of high arsenic aquifers in the central Yangtze River Basin since the Last Glacial Maximum

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ABSTRACT: Understanding the mechanism of arsenic mobilization from sediments to groundwater is important for drinking water supply and water quality management in endemic arsenicosis areas, such as the Jiangnan alluvial plain in the middle reaches of the Yangtze river. Sediment samples from three boreholes in Jiangnan Plain were collected for bulk geochemistry analysis and OSL dating. Sedimentological processes and palaeoclimatic optima after the Last Glacial Maximum have created favorable conditions for the formation of high-As aquifer systems. Bulk sediment geochemistry results indicated that As was correlated with sulfur in the pre-LGM sediments and by contrast As was correlated with iron in the post-LGM sediments. The intense chemical weathering led to sulfur depletion after the LGM could contribute to the As enrichment in the Holocene and upper Pleistocene aquifer.

1 INTRODUCTION

Geogenic enrichment of arsenic (As) in groundwater has been a topic worldwide concerned over the past decades due to its severe health threat, which impact about one hundred million people. Within river systems draining the Himalaya, tectonic movement, sedimentological processes and palaeoclimatic optima after the Last Glacial Maximum have created favorable conditions for the formation of high-As aquifer systems mainly within the Late Pleistocene-Holocene deposits (McArthur et al., 2011; Wang et al., 2017). The Jiangnan Plain located in the middle reach of the Yangtze River Basin in central China has been documented with severe As contamination in the shallow aquifers (Gan et al., 2014; Deng et al., 2018). However, the underlying mechanism responsible for the formation of high arsenic aquifers has not been clearly understood yet. This purpose of this study was to delineate the relationship between the effects of Quaternary sediment deposition responding to sea level change since the Last Glacial Maximum on the arsenic enrichment in the shallow aquifers in the Jiangnan Plain.

2 METHODS/EXPERIMENTAL

2.1 Study area

Jiangnan alluvial plain in Central China is located in the middle Reaches of the Yangtze River, formed by Yangtze River and its largest tributary the Han River (Fig. 1). It is well known as the beautiful and rich land of fish and rice. It has a sub-tropical monsoonal climate with an annual precipitation and evaporation of approximately 1200 mm and 1378 mm, respectively.

Figure 1. The geographical location of sampling borehole JH002 in the Jiangnan Plain.

The Quaternary groundwater systems can be divided into three groups of aquifers. The first group is composed of Holocene alluvial and lacustrine deposits (clay, silt and fine sand) with a thickness of 3–10 m. The second group is composed of late and middle Pleistocene alluvial sediments (medium-coarse sand and gravel, interlaced clay lenses in local area) with a thickness of 30 m; this is the main aquifer for water supply. The third group is composed of early Pleistocene alluvial sediments (fine-medium sand, interlaced clay lenses).

2.2 Sampling and analysis

Sediment samples from three boreholes with depths up to 230 m in typical arsenic-affected area were collected for bulk geochemistry analysis and As, Fe and S speciation. Major-element analysis (SiO_2 , Fe_2O_3 , Al_2O_3 , Na_2O , CaO , P_2O_5 , and MgO) was done with an XRF spectrometer (RIX2100, Rigaku). Trace elements were determined using ICP-MS after a mixed acid ($\text{HNO}_3\text{--HClO}_4\text{--HF}$) digestion. Total carbon and sulfur, total organic carbon were measured using an Elemental Analyzer (Vario MICRO cube, Elementar).

Figure 2. The relationship between arsenic and iron & sulfur in pre-LGM and post-LGM sediments samples of JH002.

We used the optical stimulated luminescence (OSL) dating method to measure the age of 23 sediment samples with the depths from 3.25 to 45.3 m below ground surface (bgs).

3 RESULTS AND DISCUSSION

3.1 Lithology and bulk geochemistry

According to the lithology and grain size analysis, the Quaternary aquifers could be divided into the Holocene-upper Pleistocene phreatic aquifer (15–60 m) and middle-lower Pleistocene confined aquifer (>60 m). Bulk sediment geochemistry analysis showed that As, Fe, Mn, P, TOC contents in the sediments of middle-lower Pleistocene aquifer were much higher than those in the Holocene and upper Pleistocene aquifer. Arsenic sequential extraction and SEM-EDS analysis indicated that the shallow sediments possessed an average As content of $9 \mu\text{g g}^{-1}$. Arsenic content in the groundwater was up to $2330 \mu\text{g L}^{-1}$. The deep aquifer possessed an average As content of $55 \mu\text{g g}^{-1}$, and the highest As content in the groundwater was about $100 \mu\text{g L}^{-1}$. Arsenic was correlated with sulfur in the pre-LGM sediments ($R = 0.7$, $P < 0.05$) and the pyrite was observed to be the main sink of As in the deep aquifer (>60 m), by contrast arsenic was correlated with iron in the post-LGM sediments ($R = 0.67$, $P < 0.05$) (Fig. 2).

Iron sequential extraction results indicated that Fe mainly existed as iron-oxides in Holocene and upper Pleistocene sediments, while the siderite was the main form of in middle-lower Pleistocene sediments. In addition, abundant Fe in pyrite-form were observed in some sulfur riched samples in middle-lower Pleistocene sediments.

3.2 Sedimentological controls on the enrichment of arsenic in shallow aquifers since the LGM

Sea level in the Last Glacial Maximum was 120 m lower than today, which led to depositional break during 40–20 ka. In warming period, the sea level raised immediately, with a quick response of the Yangtze

Figure 3. The variation of particle size, CIA index and As/S mole ratio in sediment samples of JH002 borehole.

river. The high CIA, Rb/Sr and V/Cr values indicated an intense chemical weathering and oxidizing sedimentary environment in post-LGM period. The mole ratio of As/S in the post-LGM sediments was >0.01 , while in the deep pre-LGM sediments, the As/S kept stable and ranged between 0.0032 and 0.0061 (Fig. 3). Arsenic enriched in the shallow aquifer during post-LGM period may be caused by the intense chemical weathering of arsenopyrite in the sediments after the LGM.

4 CONCLUSIONS

The bulk sediments geochemistry indicated that As was correlated with sulfur in the pre-LGM sediments and by contrast arsenic was correlated with iron in the post-LGM sediments. The intense chemical weathering leading to sulfur depletion after the Last Glacial Maximum could contribute to the As enrichment in the Holocene and upper Pleistocene aquifer.

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