Provenance and paleoenvironment impact on arsenic accumulation in aquifer sediments from the Datong Basin, China: implications from element geochemistry

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Abstract

High As aquifer sediments from the Datong Basin, China have been analyzed to indentify the provenance and depositional environment related to As accumulation in the sediments. Evidence from discrimination diagrams show that the sediments were derived from typical continental sources. Geochemical signatures suggest that the provenance of the sediments remained similar throughout the depositional period. In contrast, the distribution of Eu/Eu* ratios along the borehole requires an obvious change in the paleoenvironment. The co-variation of As contents and Rb/Ti ratios along the borehole suggests that the accumulation of As in sediments is mainly controlled by the paleoenvironment.

Keywords: Geochemistry; Provenance; Paleoenvironment; Arsenic; Datong Basin.

1. Introduction

Arsenic (As) contamination of groundwater in sedimentary aquifers has been extensively reported. Natural occurrences of As-contaminated groundwater have caused severe associated health problem. In order to eliminate this threat, it is imperative to understand the factors causing the enrichment of As in groundwater. Initially, As is regard present as solid phase within the aquifer sediments. Water-sediment interaction consequently promotes As mobilization and enrichment in groundwater. Our recent study [1] indicated that As concentration in groundwater is strongly depend on the source of aquifer sediments and

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their As contents. Therefore, in order to elucidate the generation of high As groundwater, it is important to understand the As accumulation in aquifer sediments.

Significant improvement has been made in geochemical approaches to monitoring sediment provenance, especially through the introduction of discrimination diagrams based on the relationship of major and trace elements. Recent research has suggested that sedimentary trace element records in sediments give additional information on the sediment depositional environments related to paleoclimate [2]. Therefore, to shed light on As-accumulation in aquifer sediments and its origin ascribed to a paleoenvironment, we present major and trace elements of the bulk sediments collected from a known As-contaminated site in the Datong Basin.

2. Materials and methodology

2.1. Geological setting of study area

Datong basin is located in the northern part of the China and bounded by Hengshan Mountains, Guancen Mountains and Hongshou Mountains in the southeast, west and northwest of the basin, respectively (Fig. 1). Hengshan Mountains mainly consist of Neoarchaean Hengshan Complex. Lower Paleozoic limestone outcrops in the Guancen Mountains. The Hongshou Mountains are primarily Permian to Cretaceous clastic rocks. The basin was filled with the Quaternary young sediments.

2.2. Sampling and sample analysis

Total of sixty-nine sediment samples were collected from a known As-contaminated site at the Datong Basin (Fig. 1). Core samples were taken every 1.5 to 2 m down to a depth of about 130 m. The lithologies of sediments are mainly clay and silt (Fig. 1). Major oxides compositions of the samples were determined using X-ray Fluorescence (XRF). Trace element including As analyses were carried out using a PE Elan DCR-e inductively coupled plasma mass spectrometry (ICP-MS). The C contents of the acid-leached samples (TOC) were measured on an Elementar CHN-O Analyzer. The analytical precision is better than 5% for the major elements and 5–10% for trace elements.

3. Results and discussion
3.1. Bulk geochemistry of sediment

Apart from a few sediments, the SiO\textsubscript{2} contents of sample from the Datong Basin range from 50 to 70% and are enriched relative to post-Archean Australian Shale (PAAS) [3]. The Al\textsubscript{2}O\textsubscript{3} contents of the samples are all lower than of PAAS (18.9 wt.%), in contrast, the CaO contents are very high and remarkably higher than that of PAAS (1.3 wt.%). Most samples show that the Na\textsubscript{2}O and K\textsubscript{2}O contents are slightly enriched relative to PAAS, with average values of 1.55 wt.% and 2.26 wt.%. The (La/Yb)\textsubscript{N} ratios of samples range from 0.68 to 1.09 and Eu/Eu* ratios change between 0.72 and 0.98 showing a slightly negative Eu anomalies. The Eu/Eu* ratios show significant variations in values at the depth of 20 and 90m (Fig. 2) probably related to the change in depositional environment. The strong reducing environment can cause the negative Eu anomalies in sediment samples. The Sr contents of most samples range from 100 to 500 mg/kg. The high Sr contents detected in some samples may be due to the occurrence of carbonate mineral phases, which can also result in the high CaO contents in the sediments. The Rb contents of samples range from 60.8 to 131 mg/kg and exhibit random with strata (Fig. 2).

3.2. The provenance of high As sediments

In the discrimination diagram for sedimentary provenance, almost all samples plot in the field of quartzose sedimentary provenance (Fig. 3) suggesting an obvious sedimentary recycling. Trace elements can give more reliable information about provenance and weathering. Both heavy minerals and clays are important hosts for REEs, Y, and Th in sedimentary rocks. These minerals are often considered to be resistant to chemical weathering in sedimentary rocks [4]. The La/Ti, Yb/Ti, and Y/Ti ratios of the samples from the Datong Basin show similar distribution along the borehole with little value variations indicating they are included in stable minerals relatively unaffected by weathering. Altogether the evidence from the discrimination diagrams for sedimentary provenance show that the Datong Basin sediments were derived from typical continental sources. Importantly, geochemical signatures (Eu/Eu*, La/Ti, Yb/Ti, and Y/Ti ratios) do not significant change along the borehole, suggesting the provenance of the Datong Basin remained similar throughout the depositional period.

Rb-bearing minerals are instability during chemical weathering and then Rb is readily to be leached during this process. However most of the dissolved Rb may be absorbed and fixed in secondary minerals,
which explains the fact that weathering products generally have higher Rb concentrations than parental bedrocks [4]. The variation of Rb/Ti ratios for samples from the Datong Basin may be related to: (1) change of sediment source area, (2) change of chemical weathering in source area, and (3) grain-size sorting during sediment transport. As discussed above, there are no obvious changes in sediment source areas during the depositional periods. There is also no evidence to suggest hydraulic sorting effects significantly influence elemental ratios as most samples are clay and silt. This leaves chemical weathering as the most likely explanation for the observed changes in Rb/Ti ratios and element records.

3.3. The impact of provenance and paleoenvironment on As accumulation in sediments

The geochemical discrimination diagram for sedimentary provenance indicates the similar provenance for high As sediments. The contents of As in sediments have not show significant correlation with the provenance indicators including Y/Ti, La/Ti and Yb/Ti suggesting the provenance cannot determine the contents of As in sediments. In contrast, similar variation of As contents and Rb/Ti ratios of sediments along the borehole. Therefore, the observed close correlation between As contents and Rb/Ti ratios indicates that their respective mineral host instability to weathering processes occurring at diverse ages can have significant role on As contents in sediments.

4. Conclusions

La/Ti, Yb/Ti and Y/Ti ratios of the sediments from the Datong Basin indicate no significant variations in sediments provenances during the depositional period. Rb/Ti ratios and Eu/Eu* values of the sediments therefore infer an obvious change in paleoenvironment. The similar distribution of As and Rb/Ti along the borehole suggests the accumulation of As in sediments is mainly controlled by the paleoenvironment.

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