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Provenance, chemical weathering, and sedimentary environment of the aquifer sediments: Implication for arsenic enrichment in groundwater

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ABSTRACT

High arsenic (As) groundwater has been widely observed in the world, but the provenance, chemical weathering, and sedimentary environment of aquifer sediments hosting high As groundwater remain poorly understood. The geochemical and isotopic characteristics of high-resolution aquifer sediments were investigated to unravel the roles of provenance, chemical weathering, and sedimentary environment of the aquifer sediments. The average values of rare earth elements (REEs) showed the enrichment of light REEs and flat heavy REEs in the aquifer and the Yellow River sediments. The chemical index of alteration reflected incipient and moderate weathering (41 to 71). The $\delta^{13}C_{SOM}$ (−27.4‰ ± 4.3‰) and C/N values (8.1 ± 5.8) indicated aquatic environment of sedimentation. Shallow aquifer sediments mostly had relatively high ΣREEs contents and negative Eu anomalies, demonstrating that the aquifer sediments were mainly derived from the Yellow River (up to 94.3% ± 5%). Deep aquifer sediments had relatively low ΣREEs contents and positive Eu anomalies, which originated from the Yinshan Mountains (up to 72.2% ± 20.4%) principally with igneous and metamorphic rocks. Moreover, recycling of the Yellow River sediments from Northeastern Tibetan Plateau enhanced the silicate weathering and increased contents of Fe/Mn oxides and As. The $\delta^{13}C_{SOM}$ and C/N values evidenced that organisms of paleo-lake or paleo-Yellow River were responsible for abundant sedimentary organic matter. Concentrations of Fe, Mn, and As were high in shallow groundwater from the plain. The As enrichment in the shallow groundwater was jointly controlled by the provenance and chemical weathering of the aquifer sediments (14%), sedimentary environment (10%), reductive dissolution (35%) of Fe/Mn oxides, As desorption under elevated pH (20%), and competitive adsorption of HCO_3^- (21%). This study systematically reveals the roles of provenance, chemical weathering, and sedimentary environment of the aquifer sediments and highlights joint contributions of geochemical processes and geochemical conditions to As enrichment in groundwater.

1. Introduction

High arsenic (As) groundwater is a widespread problem around the world, potentially negatively affecting several million people (Podgorski and Berg, 2020) with symptoms of blackfoot disease (Tseng et al., 1995), cardiovascular disease (States et al., 2009), and cancer (WHO, 2022). Therefore, As concentrations in drinking water has been set to less than 10 μg/L (WHO, 2022). The genesis of high As groundwater was intensively investigated to predict and locate safe groundwater (Glodowska et al., 2020; Guo et al., 2013; Postma et al., 2007). Arsenic concentrations in groundwater were controlled by reductive dissolution of bearing-As Fe/Mn oxides (Nickson et al., 1998; Smedley and Kinniburgh, 2002), oxidation of bearing-As pyrite (Jones and Pichler, 2007), and desorption (Nicolli et al., 2010). Overall, As in the aquifer sediments was believed to be the source of groundwater As (Shen et al., 2018; Stuckey et al., 2015).

Distributions of high As groundwater, orogenic belts, and foreland basins in the world illustrated that high As groundwaters were mostly

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