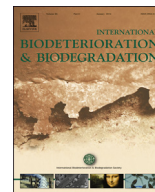




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Arsenic in geothermal systems of Tengchong, China: Potential contamination on freshwater resources

Zhou Jiang^{a, b}, Ping Li^{a, *}, Jin Tu^a, Dazhun Wei^a, Rui Zhang^a, Yanhong Wang^b, Xinyue Dai^{a, b}^a State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, PR China^b School of Environmental Studies, China University of Geosciences, Wuhan 430074, PR China

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ABSTRACT

To detect arsenic geochemistry and evaluate its environmental risk, 30 geothermal water samples were collected from Rehai and Ruidian geothermal fields of Tengchong, China. There were three main water types including SO₄ and Na-Cl-HCO₃ types in Rehai and Na-HCO₃ type in Ruidian. Arsenic concentrations ranged from 22.1 to 1150.3 µg L⁻¹. Relative high concentrations of arsenic with an average of 495.1 µg L⁻¹, as well as K, Na, F, Cl, Li and B, were detected in neutral or alkaline geothermal water samples, which suggested that rock leaching in geothermal reservoir was the main resource of arsenic enrichment in geothermal water. Comparatively, those acid geothermal water samples which were formed by vapor rich in H₂S condensing and oxidizing in the shallow groundwater or surface water had lower arsenic concentrations with an average of 77.1 µg L⁻¹. The ratios of arsenite to total arsenic widely ranged from 0.01 to 0.97 and were determined by mixing proportions of geothermal water and shallow groundwater or surface water. Furthermore, there was a significantly positive correlation between arsenic with HCO₃ (R = 0.81, P = 0.01). These results implied that shallow groundwater or surface water in Tengchong geothermal area had been probably contaminated by arsenic due to the mixing of geothermal water.

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1. Introduction

Arsenic (As), notoriously known as a toxic and carcinogenic metalloid, is a ubiquitous trace component in geothermal system (Feng et al., 2014; Keller et al., 2014; Maizel et al., 2016). It occurs together with other environmental contaminants such as fluoride (F), lithium (Li), boron (B) and antimony (Sb), recognized as being a typical “geothermal suite” of contaminants (Webster and Nordstrom, 2003; Kaasalainen and Stefánsson, 2012; Shah et al., 2015). The occurrence of As in terrestrial geothermal systems has been identified in many countries of the world, including USA, Canada, Iceland, New Zealand, Philippines, Japan, Russia, Turkey, France, Spain, China and Latin America (Smedley and Kinniburgh, 2002; López et al., 2012).

In geothermal reservoir, reducing condition prevails and As is generally present as arsenite (AsIII) (Farnfield et al., 2012). Due to several physical and chemical processes during their ascent from

the geothermal reservoir to the ground surface, geothermal waters change their chemical composition so that eventually form four main water types including mature Na-Cl water, acid SO₄ water low in Cl, acid SO₄-Cl water and HCO₃-rich water (Kaasalainen and Stefánsson, 2012; López et al., 2012). High As concentrations are often found in mature Na-Cl water and acid SO₄-Cl water (D'Imperio et al., 2007; Birkle et al., 2010). Though AsIII is the predominant species in geothermal source water, arsenate (AsV) can also be present, with AsV/AsIII varying due to mixing with shallow oxygenous groundwater or surface water and subsequent oxidizing (Donahoe-Christiansen et al., 2004; Bahar et al., 2016). Arsenic in geothermal areas could contaminate adjacent surface water and groundwater resources with its mobilization and transformation in geothermal setting under changes in geochemical and microbiological conditions, which have attracted more attention by researchers (Aksoy et al., 2009; Piqué et al., 2010; Ilgen et al., 2011; Bundschuh et al., 2013; Li et al., 2014).

Tengchong, located in southwestern of China, is a typical volcanic geothermal area and has abundant geothermal resources (Guo, 2012). Hot springs in this geothermal area have been found with As concentrations up to 850 µg L⁻¹ (Guo and Wang, 2012).

* Corresponding author.

E-mail address: pli@cug.edu.cn (P. Li).

Over the last decade, mineralogical, hydrogeochemical and microbial studies have been performed in Tengchong geothermal area, based on which it was found that these hot springs had diverse chemical characteristics and the deep geothermal liquid might be mixed with groundwater or surface water during ascending (Zhang et al., 2008; Guo and Wang, 2012; Song et al., 2013; Hou et al., 2013; Briggs et al., 2014). However, so far, As geochemistry in geothermal water and its potential environment risk in this geothermal area have yet to be studied systematically. Therefore, this study was aimed to investigate As geochemical behavior and characteristics in geothermal water of Tengchong geothermal area, and then evaluate the potential As contamination on freshwater resources.

2. Materials and methods

2.1. Site description

Tengchong geothermal area is located in Yunnan Province of southwestern China and near the border between China and Myanmar. Tectonically, it lies at the eastern end of the Yunnan-Tibet geothermal belt, a major part of the Mediterranean-Himalayas geothermal belt (Qin et al., 2011). The topography of Tengchong is high in the north and low in the south, with the range of altitudes from 930 m to 3780 m (Guo and Wang, 2012). There are three major rivers crossing in Tengchong including Binglang River, Daying River and Longchuan River. Tengchong has a subtropical monsoon climate with an annual mean temperature of 14.9 °C and an average annual rainfall of 1480 mm, with 85% of it from June to October (Jones and Peng, 2012).

Tengchong geothermal area, formed during the Cenozoic as the Burmese Block was thrust under the Tengchong Microplate, is characterized by numerous volcanoes and extensive faulting (Shangguan et al., 2005). It has wide distribution of volcanic rocks including basalt, andesite basalt, andesite, and dacite, with an outcropping area up to 1000 km². There are more than 50 volcanoes and 140 geothermal fields throughout Tengchong County. Rehai and Ruidian geothermal fields are the two largest hydrothermal areas in this region. The Rehai geothermal field, located 11 km southwest of Tengchong County, has a total area of about 10 km² with the highest inferred reservoir temperature (69 °C–450 °C) in Yunnan (Du et al., 2005). With increasing depth, the strata beneath the Quaternary volcanic rocks successively consist primarily of Holocene alluvium, Middle Pleistocene basalt, Lower Pleistocene andesite, Miocene brecciated rocks, Cretaceous granite and Proterozoic gneiss and migmatite (Guo and Wang, 2012). There is a wide distribution of faults including main nearly N-S stretching and secondary NE-SW and NW-SE stretching in this field, which serve as the channels for the upward flow of geothermal fluids and feed to abundant surface hydrothermal manifestations including boiling springs, hot springs, intermittent spouting springs, hydrothermal explosions, fumaroles, steaming ground (Fig. 1) (Guo and Wang, 2012). The Ruidian geothermal field was located 39 km north of Tengchong County, with an area of about 0.6 km². The strata assemblage in this field is composed of granites, sandstones and basalts (Zhang et al., 2008). Generally, the hydrothermal activities in Ruidian are less strongly as those at Rehai, no matter their quantity, type and active degree of hot springs.

2.2. Sample collection and preservation

Twenty-three samples from the Rehai geothermal field and seven samples from the Ruidian geothermal field were collected in May 2013 (Fig. 1). The sinter of sample Zhenzhuquan (RH15-RH-17) was collected with spatulas and spoons, and placed into 50 mL sterile polypropylene tube for determining total As (As_{Tot})

concentrations. All water samples were filtered through 0.22 µm membranes on site into 50 mL acid-washed polyethylene bottles which had been rinsed with filtered water twice before sampling. For cation analysis, the samples were preserved with a few drops of concentrated HNO₃ and bring the pH below 1. Temperature, pH, electrical conductivity (EC) and dissolved oxygen (DO) were measured in the field with hand-held meters that was calibrated prior to sampling. Alkalinity was determined using the Gran titration method (Guo and Wang, 2012) on the same day as the sample was collected. As species separation was done in situ, following the method reported by Le et al. (2000). Briefly, 10 mL of each water sample was passed through a silica-based strong anion-exchange cartridge (Supelco). AsV was adsorbed and AsIII remained in the filtrate. Subsequently, the anion-exchange cartridge was eluted with 1 M hydrochloric acid, and the eluent was analyzed for AsV.

2.3. Hydrochemical and statistical analyses

The cation and anion concentrations were measured by inductively coupled plasma-optical emission spectrometry (ICP-OES, CAP6300, Thermo) and ion chromatography (ICS1100, Dionex), respectively. AsIII and AsV concentrations were determined using hydride generation atomic fluorescence spectrometry (AFS-9600, Haiguang) according to Jiang et al. (2014), with the detection limits for AsIII and AsV of 2 and 4 ppb, respectively. As_{Tot} in the sinter from sample Zhenzhuquan was extracted by 1:1 aqua regia digestion method in a water bath (Deng et al., 2011; Gong et al., 2015) and measured by AFS-9600. Principal component analysis (PCA), hierarchical cluster analysis, correlation analysis and scatter plots between the physico-chemical parameters and As concentrations or the AsIII/As_{Tot} were performed using R software version 3.1.1 (<http://cran.r-project.org>) (Liang et al., 2014). Samples were hierarchical clustered based on all physico-chemical parameters using unweighted pair-group method with arithmetic means (UPGMA) and Euclidean distance measure. Piper diagram was prepared using the RockWare Aq•QA 1.1.1.

3. Results and discussion

3.1. Chemical characteristics of geothermal water

In Rehai geothermal field, geothermal water samples had wide ranges in pH (2.05–8.87), temperature (34.7–96.6 °C), EC (540–5310 µS cm⁻¹) and DO (0–3.19 mg L⁻¹) (Table 1). The water types of those acid geothermal water samples were diverse, with SO₄ as predominant anion and K, Na, Ca as alternative predominant cation, whereas Na-Cl-HCO₃ was the main in alkaline geothermal water samples (Table 1; Fig. 2). Different from Rehai, all the geothermal water from Ruidian geothermal field were closely neutral with pH from 6.55 to 7.75, temperature from 58.3 to 80.0 °C, EC from 3390 to 4830 µS cm⁻¹ and DO from 0.19 to 0.43 mg L⁻¹, and presented in a sole Na-HCO₃ type (Table 1; Fig. 2). PCA result showed that these geothermal water samples from Rehai geothermal field were generally classified into three groups (Fig. 3). Group 1, located in the second and third quadrant, is composed of all acid geothermal water samples characterized with high concentrations of SO₄, Fe, Al, Zn and Mn. Group 2 was located in the first quadrant, clustered by some alkaline geothermal water with high concentrations of Ba and Sr and moderate concentrations of K, Na, F, Cl, Li, B and Se. In fourth quadrant, group3 was dominated by some other alkaline geothermal water samples with high K, Na, F, Cl, Li, B and Se concentrations. Closely neutral samples from Ruidian geothermal field were generally similar to the second group, but had much higher concentrations of Ca, Mg, Ba and Sr. These groupings were consistent with the hierarchical cluster analysis

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