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Geochemical genesis of arsenic in the geothermal waters from the Rehai hydrothermal system, southwestern China

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Abstract

The geothermal waters from the Rehai magmatic hydrothermal system, located in the Tengchong volcanic region, south-western China, are characterized by diversified hydrochemical types and a wide range of pH values. The neutral hot springs in Rehai generally have much higher arsenic concentrations than the acid hot springs. Further inspection shows that the acid, sulfate-rich springs are essentially locally-perched groundwaters heated by H_2S-rich steam separated from deep geothermal fluid. Hence, the dissolution of near-surface rocks at moderate temperatures is the sole source of arsenic in the steam-heated acid springs. In comparison, the arsenic in the neutral hot springs, which represent the deep-circulating geothermal fluids, comes from long-term water-rock interactions at much higher reservoir temperatures. Moreover, the possibility that magmatic fluid is a potential arsenic source for the neutral springs in Rehai cannot be ruled out.
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1. Introduction

Arsenic (As) enrichment in natural waters is of great concern because of chronic arsenic poisoning related to drinking water sources. In recent years, arsenic from hydrothermal systems is attracting more and more attention. The arsenic concentrations of geothermal fluids from most hydrothermal systems across the world are usually one to three orders of magnitude greater than those of unpolluted cold groundwaters. In China, hydrothermal areas are found in almost every province. However, high-temperature hydrothermal systems where high-arsenic geothermal

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waters occur are found mainly in the Tibet autonomous region, western Yunnan Province and western Sichuan Province, i.e. the so-called Yunnan-Sichuan-Tibet Geothermal Province (YST).

Across the YST Geothermal Province, the Rehai hydrothermal area, located in the Tengchong volcanic region of Yunnan Province, is geothermally unique because it is the only hydrothermal system in mainland China discharging a large number of acidic springs with pH as low as 2. The arsenic concentrations of the Rehai geothermal waters are also high (the highest is up to 1.1 mg/L). The diversified hydrochemical types, the wide range of pHs and the high arsenic concentrations of the geothermal waters from Rehai offer a precious opportunity to comprehensively investigate the geochemical genesis of arsenic in geothermal water. Thus, the primary aims of this study are to characterize the arsenic geochemistry of the Rehai geothermal waters and to clarify its geochemical origins.

2. Background geology, sampling and analysis

Rehai is the largest and most active hydrothermal area in Tengchong, and its tectonic background and stratigraphic sequence were described in detail by Guo and Wang (2012)[1]. The Rehai hydrothermal system is hosted by Yanshanian granite and Proterozoic metamorphic rocks, whereas its cap rocks consist of intensively altered Miocene clastic rocks. A number of geological, geophysical and geochemical studies have demonstrated that there is a magma chamber below Rehai and it was possibly formed during the fourth stage of Tengchong magmatism, i.e. the Middle-Late Pleistocene.

Both neutral and acidic springs at Rehai were sampled for full hydrochemical analyses. The samples were filtered through 0.45 μm membranes on site and pre-treated according to the requirements for various chemical analyses. Water temperature and pH, as unstable parameters, were measured with hand-held meters that were calibrated prior to sampling, and H_2S was determined in situ using a HACH colorimeter. Total alkalinity was determined using the Gran titration method on the sampling day, based on which the HCO_3^-, CO_3^{2-} and CO_2 concentrations of all samples were calculated using PHREEQC. The concentrations of cations, anions and trace elements were analyzed by ICP-OES, IC and ICP-MS within 2 weeks after sampling, respectively.

3. Results and discussion

3.1. Hydrochemical characteristics

All the geothermal water samples collected in the Rehai geothermal field can be clearly divided into two groups based on their arsenic concentrations: one with arsenic concentrations ranging from 0.31 to 1.10 mg/L is also characterized by high pH values (7.61 - 8.87), very high Cl concentrations (362 - 725 mg/L), and low SO_4 concentrations (19 - 38 mg/L), and the other with lower arsenic concentrations (from 0.01 to 0.21 mg/L) has low pH values (2.05 - 6.82), low Cl concentrations (6 - 77 mg/L), and high SO_4 concentrations (128 - 2134 mg/L). That is, the water in the neutral hot springs in Rehai has much higher arsenic concentrations than that in the acid hot springs. Moreover, the acid spring waters also have much lower B, F, Li, Rb, Cs, and Na concentrations. However, their Ca, Mg, Fe, and Al concentrations are much higher than those of the neutral springs.

3.2. Geological genesis of the Rehai hot springs

Across the world, three different types of hot springs are usually identified in a typical volcanic-magmatic-hydrothermal system: acid SO_4-Cl (or Cl-SO_4) springs, neutral Cl springs and slightly alkaline to neutral HCO_3-Cl springs. Below such a system, the cooling magma chamber is capable of continuously releasing a magmatic fluid composed primarily of water and acid gases (such as SO_2, HCl, HF, H_2S and CO_2) with a potential to form highly immature waters characterized by very high SO_4 concentration, comparatively high Cl concentration and very low pH value. For some hydrothermal systems with a shallow magmatic heat source connected to supergene groundwater by fractures, portions of the magmatic vapor are able to ascend to very shallow levels and are absorbed directly into local groundwaters[2,3]. In this case, high-temperature, acid, SO_4-Cl or Cl-SO_4 springs are likely to form because the discharged geothermal waters seldom have sufficient time to be fully neutralized via reaction with subsurface rocks and considerable magmatic sulfur remains in geothermal water. However, if the magma chamber

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