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Research paper

## Hydrogeochemical reconnaissance of arsenic cycling and possible environmental risk in hydrothermal systems of Taiwan



Jyoti Prakash Maity<sup>a,b</sup>, Chien-Yen Chen<sup>a,\*</sup>, Jochen Bundschuh<sup>c,d</sup>, Prosun Bhattacharya<sup>b,d</sup>, Abhijit Mukherjee<sup>e</sup>, Young-Fo Chang<sup>a</sup>

<sup>a</sup> Department of Earth and Environmental Sciences, National Chung Cheng University, 168 University Road, Ming-Shung, Chiayi County 62102, Taiwan

<sup>b</sup> School of Civil Engineering and Surveying & International Centre for Applied Climate Science, University of Southern Queensland, Toowoomba, Australia

<sup>c</sup> Deputy Vice Chancellor's Office (Research and Innovation) & Faculty of Health, Engineering and Sciences, University of Southern Queensland, Toowoomba, Australia

<sup>d</sup> KTH-International Groundwater Arsenic Research Group, Department of Sustainable Development, Environmental Science and Engineering, KTH Royal Institute of

Technology, Teknikringen 76, SE-10044 Stockholm, Sweden

e Department of Geology and Geophysics, Indian Institute of Technology (IIT)-Kharagpur, Kharagpur 721302, West Bengal, India

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#### ABSTRACT

Hydrothermal activity creates geo-hydro-chemical interactions between hot water/fluid and the host rocks, which changes the hydro-chemical composition of the geothermal water/fluid and enriches trace elements. Existence of arsenic (As) is reported from different hydrothermal systems as well as several region in groundwater system at elevated concentration globally, compared to 10 µg/L WHO (World health Organization) guideline. The distribution of dissolved major and minor elements, including arsenic (As) was studied in hydrothermal systems of Taiwan. For the first time in Taiwan As(V) and As(III) species were researched from the three principal geological settings of Taiwan. Aim was to understand the cycling, fate and transport and potential impact of As on the surficial hydrological systems. Water samples were collected from sixteen hydrothermal springs of 3 different geological settings. Three groups of hydrothermal spring water samples could be distinguished: (i) strongly acidic (pH < 3), sulfate-enriched waters of H-SO<sub>4</sub>-type (Yangmingshan, and Taipu, Beitou), (ii) slightly alkaline waters (pH: 8-8.95) (Jiben, Antung and Kung-Tzu-Ling), and (iii) circum-neutral waters (pH 6.47-7.41) of Na-HCO<sub>3</sub>/Na-Cl-HCO<sub>3</sub>-type (Wulai, Hongye, Rueisuei, Chung-Lun and Biolai). The waters are enriched with alkali and alkali earth metals compared to drinking water. Similarly, the water of most of the geothermal springs were found to be enriched with As (highest concentration at Beitou: 1.456 mg/L) with As(III) being the principal As species. Arsenic concentrations of hydrothermal spring waters in igneous rock terrains exhibit highest concentrations  $(0.69 \pm 0.71 \text{ mg/L})$  followed by those of sedimentary (0.16  $\pm$  0.14 mg/L) and metamorphic (0.06  $\pm$  0.02 mg/L) terrains. The discharged geothermal springs water contaminate the surface and groundwater (including drinking and irrigation water resources), where significant levels of arsenic and other toxic element have detected and hence being a significant risk for human health and environmental.

#### 1. Introduction

Hydrothermal systems, with geothermal fluids discharging as springs, fumaroles or steam at or close to the surface of the earth are found at several places of the world (Gemici and Tarcan, 2004; Baba and Ármannsson, 2006; Brown and Simmons, 2003; Bundschuh et al., 2017; Maity et al., 2011a). These hydrothermal springs are related to local or global tectonism, and can be attributed to three general geological settings (Webster and Nordstrom, 2003). These include (i) the subduction zones where marine crust is being subducted below continental crust such as the Himalyan Indus-Tsangpo convergent zone (India, Nepal and Tibet), the convergent plate margins of the Pacific (e.g., geothermal fields in New Zealand, Papua New Guinea, Philippines, Indonesia, Japan, Kamchatka, Alaska, western USA, Mexico, Chile, Costa Rica, Argentina, etc.); (ii) the Hot Spots, resulting from local magma chambers developed at shallow depth within the asthenosphere (e.g., Azores, Hawaii, and western USA such as Yellowstone); and (iii) the rift zones, i.e., divergent plate boundaries (e.g. Turkey (Bundschuh et al., 2013), Gregory Rift Valley in the Eastern African Rift, affecting Ethiopia, Kenya and Tanzania (Bhattacharya et al., 2015;

\* Corresponding author.

E-mail address: chien-yen.chen@oriel.oxon.org (C.-Y. Chen).

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Fig. 1. Schematic diagram is showing the plate tectonic setting and associated major submarine physiographic features in Taiwan hydrothermal areas (modified from MOEA, 2017).

Ligate et al., 2016), the Rio Grande Rift valley in Colorado and New Mexico of the southern USA, and the rift zone systems of Iceland) (Webster and Nordstrom, 2003). These hydrothermal systems show various specific hydrochemical characteristic, with suites of enriched (compared to drinking water) trace elements [arsenic (As), antimony (Sb), boron (B), fluoride (F), lithium (Li), mercury (Hg), selenium (Se), thallium (Tl)], which are considered common contaminants in surface hydrological and groundwater systems (Webster and Nordstrom, 2003; Bundschuh and Maity, 2015; Bundschuh et al., 2017).

Arsenic is present at elevated concentrations, compared to average concentration of As in groundwater, in most of the hydrothermal systems throughout the world. Because of its high toxicity, As concentrations in water exceeding the 10 µg/L WHO guideline value (WHO, 1993), are a major concern to large global population groups. Groundwater (drinking-water wells) and surface-water in the southern part of the Poopó Lake basin, Bolivian Altiplano, exceed the As concentration WHO guideline value of 10 µg/L (Ormachea Muñoz et al., 2015, 2016). Arsenic-enriched water emerging from hydrothermal springs impact downstream surface and groundwater mixing including related hydro (bio)geochemical processes, which are further influenced by respective changes of temperature, pressure, redox and pH conditions (Mukherjee and Fryar, 2008). These impacts of geothermal water on freshwater resources can significantly degrade these resources making them unsuitable for drinking or irrigation without further treatment. Hydrothermal springs in the eastern edge of the Pacific plate like the hydrothermal springs of New Zealand (e.g. Wairakei, Waiotapu, Ohaaki, Broadlands), and Mt Apo in Philippines have been reported to contain As concentrations of up to 6.2 mg/L (Ritchie, 1961; Ellis and Mahon, 1977; Webster, 1999, 1990; Guo et al., 2007). However, significantly higher As concentrations have been recorded in the western edge of the Pacific plate, e.g. the hydrothermal systems of El Tatio (northern Chile), Copper River (USA), Yellowstone National Park (USA), and Los Humeros (Mexico), 75 mg/L (Ball et al., 1998; Romero et al., 2003; Motyka et al., 1998; Ellis and Mahon, 1967; Gonzalez-Partida et al., 2001). Hence, it is understandable, that water from

hydrothermal springs, even if attributed to the same geological/tectonic setting, can still have very different concentrations of As and trace element compositions (Bundschuh and Maity, 2015; López et al., 2012; Maity et al., 2011a, 2016).

Although the North and South American hydrothermal springs have been widely studied (Webster and Nordstrom, 2003; Kulp et al., 2008), the hydrochemistry of the springs in the northeastern Pacific have not attracted much attention. Arsenic pollution in Taiwan is known for a long time (Maity et al., 2011a, 2011b; Kulp et al., 2008; Liu et al., 2009, 2011a; Nath et al., 2011), but was mostly related to marine sediments trapped within continental aquifers. The contamination of As from the over 100 known hydrothermal springs noticed in Taiwan (Maity et al., 2011a; Bundschuh and Bhattacharya, 2010; Chang et al., 2007, 1999). These hydrothermal systems include hot springs, mud springs, and seabed hot springs (Nath et al., 2011; Jang et al., 2012). During the last few decades, hot spring spas and resorts have gained enormous popularity in Taiwan. However, the enrichment of these of hot spring waters with toxic elements such as As, Se, Cr and B can have severe human health and environmental impacts and may affect the natural drinking water and irrigation water sources located at or near hydrothermal fields.

Thus, the present study is a first attempt of a reconnaissance of the water chemistry of the major and most popular hydrothermal spring systems of Taiwan being representative for different principal geological settings of the island, and evaluate their relation with their geological and tectonic setting. Such better understanding of the trace element cycling, specifically that of As, in these springs will help to better understanding the cause of As enrichment in major parts of Taiwan, along with the role of hydrothermal fluids in evolution of global hydrological systems and the related environmental risk.

## 2. Study area: geological setting and hydrogeological characteristics of hot spring in Taiwan

Taiwan (area 36,000 km<sup>2</sup>) is located at a tectonically active,

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