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ARSENIC IN GEOTHERMAL WATERS

[1]*Abioye, O. FAYIGA and [2]Uttam, K. SAHA

[1]Stockton University, NJ, USA

[2]The University of Georgia, 2300 College Station Road, Athens, GA 30602, USA

*Corresponding author

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INTRODUCTION

Geothermal waters are underground water with a temperature of at least 20 ^oC (Wiktorowicz, 2017). Geothermal energy is a form of renewable energy that makes use of heat emanating from geothermal waters to produce electricity and space heating of buildings such as greenhouses, spas and aquaculture (Baird and Cann, 2012). Deep groundwater with a temperature greater than 180 ^oC circulating within a geothermal zone and heated by contact with hot rocks is usually found in volcanic regions and island chains (Baird and Cann, 2012). Geothermal systems can either be volcanic or non-volcanic with volcanic based on the emplacement of magma (Meju, 2001). Volcanic geothermal systems include convective hydrothermal systems in hot dry rocks while non-volcanic geothermal systems involves hot fluids in sedimentary rocks flowing through fractures or faults (Meju, 2001). Volcanic rocks can be considered a heat source for geothermal waters (Yildirim and Ozgur, 2017).

In their study of geothermal waters in Turkey, Bello et al. (2017) reported that the rocks were overlain with volcanic rocks and lake sediments of Miocene were deposited. In Russia, the geothermal area is located in the volcanic belt where three groups of thermal manifestations were identified by Chudaev et al. (2017). The three different manifestations were first the fumaroles in the crater itself, secondly, the North Mutnovskaya volcanic zone and thirdly, the volcano's peripheral thermal fields (Chudaev et al., 2017). There is almost a hydrothermal system in every province in China and in southwest China, the geothermal waters are located in the Tengchong volcanic region (Guo et al., 2017). Geothermal waters are not restricted to areas with recent volcanic activities but can also be associated with regional faults that serve as channels for deep penetration of meteoric water in regions with high heat flow related and intensive tectonism (Arango-Galvan et al., 2015).

The local geology is very important in the production of geothermal waters. There are many low to moderate geothermal resources in Turkey which are related to important fracture systems (Bello et al., 2017). The Simay geothermal system in Turkey is believed to be controlled by the active Simay fault which is driven by higher than normal heat flow (Ilkisik, 1995). Bello et al (2017) explained that thermal waters escape into the tectonic zone of weakness, namely the Simay fault, as hot springs to the surface. An investigation of the Muradiye-Caldiran (Van) geothermal field in eastern Turkey revealed that most of the thermal water is discharged along NW-SE trending faults and circulation of thermal water is closely related to major faults and fracture zones (Duzen and Ozler, 2015).

Similarly, well developed faults in the Gonghe geothermal region in China acts as channels for the convective circulation of geothermal waters in the region (Liu et al., 2017). Geothermal waters in Turkey were also related to faults which are generated by compressional tectonic stresses and uplift between two extensional rift zones (Yildirim and Ozgur, 2017). Yilmaz and Ozgur (2017) explained that the geothermal waters ascend in the weakness tectonic zones at the rift zone as hot springs, steams and gases. Giordano et al. (2016) explained that the location of hot springs suggested that hot fluid occurs in areas intensely fractured.

Regardless of location of these geothermal waters, their content of toxic metalloids such as arsenic are a major concern because they may potentially contaminate groundwater which are sources of drinking water to a large population. For example, there is evidence for a mixing process between the fresh groundwater and deep geothermal waters in Babacık Pınarı in western Anatolia, Turkey (Yilmaz and Ozgur, 2017). This mixing process may contaminate fresh groundwater since arsenic concentrations of geothermal waters are about one to three times higher than those of cold groundwaters that are not polluted (Guo et al., 2017).

Exposure to arsenic is a public health concern because the World Health Organization (WHO) has declared arsenic a carcinogen which means cancer causing substance. Exposure to As has been associated with health problems such as cancer, cardiovascular and respiratory diseases, hearing problems, reproductive health problems in pregnant women and it affects the unborn (Fayiga and Saha, 2016).

A severe groundwater As contamination occurred in Bangladesh and west Bengal, India decades ago due to dissolution of arsenic rich minerals in underlying parent rocks (Ravenscroft, 2011). It was observed that symptoms of chronic As toxicity developed after 6 months to 2 years or more of exposure in India and Bangladesh (Rahman et al., 2001). Long-term exposure to As in Bangladesh produced severe arsenicosis which was revealed in skin problems, diabetes mellitus, vascular disease, neuropathy, and also multiple cancers (Ravenscroft, 2011). There was a large percentage of water samples that had high As concentrations above acceptable limits in

Argentina which led to development of skin cancer in patients with arsenicosis while there was a higher incidence rate of colorectal, lung, breast, prostate and skin cancer (Bardach et al., 2015). There are very few papers calling the attention of the public to toxicity of geothermal waters and environmental health effects. The natural sources of As in ground water are dependent on the local geology, hydrology and geochemical characteristics of the aquifers (Bhattacharya et al., 1997). This paper attempts to discuss the genesis of geothermal waters and their hydrochemical characteristics with particular emphasis on arsenic.

Hydrogeochemistry of Geothermal Waters

Thermal sprigs are main geothermal manifestations and their chemical characteristics often provide information about the origin of geothermal activity (Cruz et al., 2013). Both geochemical reactions and salt precipitation has been reported to influence the heat mining rate from geothermal waters. Cui et al. (2017) reported that the rate of heat mining decreased under the combined influence of geochemical reactions and salt precipitation. Salt precipitation has a higher effect than geochemical reactions on rate of heat mining (Cui et al., 2017). Mineral precipitation and dissolution often accompany the migration of geothermal waters (Zhang et al., 2019). Processes such as boiling, evaporation, concentration and cold-water mixing can occur as hot water from deep thermal reservoirs reach the ground (Zhang et al., 2019).

The hot springs in Indonesia indicated outflow setting with some springs classified as chloride-bicarbonate and some others classified as bicarbonate mainly. The chloride-bicarbonate indicate they came from the reservoir while the bicarbonate is either mixed with meteoric water or heated surface water (Fauziyyah et al., 2016). A study in China by Gu et al. (2017) showed how hydrogeochemical characteristics are related to different hydrogeological systems. The main type of water in twelve thermal springs evaluated were bicarbonate-sodium types with temperatures ranging from 22.3 to 41.0 ^{o}C while the mineral springs were bicarbonate-calcium-sodium waters with temperatures between 10.9 to 12.9 ^{o}C (Gu et al., 2017). Isotopic composition showed that the thermal springs were of deep circulating meteoric origin while the mineral waters were from shallow circulating waters.

Similarly, the isotopic composition of thermal waters in Turkey indicate a deeply circulating meteoric origin and geochemical characterization showed the geothermal waters is of the sodium-bicarbonate-chloride type (Gultekin et al., 2019). Hydrogeochemical evaluation of the geothermal field in Turkey showed that the temperature of the thermal waters was 36 ^{o}C while the mineralized spring was about 11 ^{o}C with mean pH given as 6.83 and electrical conductivity (EC) of 5731 μS/cm (Gultekin et al., 2019). Another study in western Anatolia, Turkey reported geothermal waters classified as sodium-chloride or sodium-chloride-bicarbonate type (Ozgur et al., 2017).

Table 1: Geochemical characteristics of selected geothermal waters

	Temp (ºC)	pH	EC (µS/cm)	Mg (mg/L)	Na (mg/L)	Cl (mg/L)	HCO_3 mg/L	Reference
Indonesia	35-45	7.0-7.6	1710-2890	37.4-227	17.4-406	8.1-465	427-2100	Deon et al. 2015
China	6-41	6.6-7.8	77-909	0.1-4.72	4.5-183	0.1-31	44-336	Gu et al. 2017
Turkey	2.9-39-3	6.2-8.5	111-6207	6.55-65.6	11.5-1612	0.83-1367	155-2691	Gultekin et al 2019
Russia	20-39	6.1-6.9	NA	17.1-45.0	140-358	7.3-33	1232-1910	Shestakova et al., 2018
UK	13.5-15.2	6.6-7.9	1900-40900	31-641	215-8550	380-11700	NA	Burnside et al., 2016
Peru	51.6-91.8	6.0-8.0	2.7-3.3	5-24	304-658	439-746	93-219	Cruz et al., 2013
Argentina	30.3-70.2	5.8-6.8	>6430	NA	NA	NA	NA	Giordano et al., 2016
Japan	40.8-75.7	1.4-3.2	NA	16.8-64.4	29.8-107	55.6-127	NA	Kikawada et al., 2017
Italy	25.2-76.0	6.2-11.6	NA	25.0-1290	32-11300	36-19348	168-1098	Montanari et al., 2017
Morocco	28-55	5.46-7.41	478-15580	16.5-247	17.2-2562	24.1-4116	192-1376	Jilali et al., 2018

Thermal waters in Russia range from the high temperature bicarbonate-sodium-calcium type waters to the lower temperature bicarbonate-sodium-calcium type waters indicating a mixing between higher Na/Ca geothermal waters and higher Na/Ca surface groundwaters (Shestakova et al., 2018). Geochemical characterization of geothermal waters in Peru indicated that the geothermal waters originated by the mixing of meteoric water and magmatic water that was circulating in sedimentary deep rocks (Cruz et al., 2013). The geothermal waters of Peru are of the alkaline-chloride-sulphate water type (Cruz et al., 2013). Isotopic data for geothermal waters from Argentina shows a predominantly meteoric origin (Giordano et al., 2016). The high EC, Mg, Na and Cl- in geothermal waters from UK (Table 1) may be due to the mine water geothermal energy production scheme in the area (Burnside et al., 2016).

Arsenic in Geothermal waters

Due to water scarcity, geothermal waters were supplied to urban communities and used for irrigation and cattle breeding in the Mexican Highlands but the arsenic concentrations measured in the waters range from 0.3 to 3.8 mg/L (Rodriguez et al., 2015). These concentrations are higher than the maximum contaminant limit of 10 µg/L set by the World Health Organization (WHO).

Table 2: Arsenic concentrations in geothermal waters

Country	As (mg/L)	Reference
Mexico	0.3-3.8 mg/L	Rodriguez et al., 2015
Turkey	680-1150 µg/l.	Esetlili et al., 2014
China	22.1-1150.3 µg L^{-1}	Jiang et al., 2018
Taiwan	0.06-1.46 mg/L	Maity et al., 2016
Slovakia	36.7 mg/L	Vranovská et al., 2015
Iceland	7-116 µg L^{-1}	Keller et al., 2014
New Zealand	0.008 to 9.08 mg l-1	Lord et al., 2012

There were a few cases of keratosis in one of the communities in Mexico where the geothermal water was supplied and some cheese producers reported low concentration of arsenic in their products (Rodriguez et al., 2015). Exposure to low level arsenic (< 50 µg/L) in a geothermal area in Italy was associated with skin diseases and circulatory system diseases (Profili et al., 2018). Geothermal features linked to bathing pools are a potential risk to human health if they have high arsenic levels (0.008 to 9.08 mg l^{-1}) such as reported in New Zealand (Lord et al., 2012). In such situations, dermal absorption is a potential route of arsenic exposure (Lord et al., 2012).

Masuda (2018) has reported that the distribution of arsenic is strongly related to areas of active plate tectonics, magmatism and associated hydrothermal activity with sources of arsenic contamination identified as mainly hydrothermal water, sulfide and arsenide minerals, volcanic ash, and iron oxyhydroxide/oxide as weathering products. High concentration of arsenic (10-126 mg/L) on geothermal waters in Tibet was attributed to contribution from the underlying magma chambers (Guo et al., 2019). The magma chambers are likely mantle derived intrusions which are severely contaminated by the deep-seated arsenic-rich sedimentary rocks.

The geological genesis of the magma fluid and its chemical composition is the most critical factor controlling the arsenic concentration of geothermal waters discharging from a magmatic hydrothermal system (Guo et al., 2019). Two sources of arsenic were identified in the volcanic geothermal fluids of Latin America; arsenic partitioned into volcanic gases and emitted in plumes and fumaroles, and arsenic in rocks of volcanic rocks that are leached by groundwaters enriched in volcanic gases (Lopez et al., 2012).

Different species of arsenic such as arsenite, arsenate and thioarsenic species exists in geothermal waters. The main species of arsenic in Daggyai geothermal water in Tibet are arsenite and arsenate though thioarsenic usually exists as the dominant species of arsenic in sulfide-rich thermal springs (Yan et al., 2019). This is based on the fact that a high concentration of sulfide can promote the changing of arsenic into thioarsenic and strong reducing environment is required for the existence of thioarsenic (Yan et al., 2019).

A study has shown that thermophilic microbes such as sulfate reducing bacteria might be involved in the formation of thioarsenates in geothermal systems (Wu et al., 2017). The concentration of arsenic in geothermal waters depends on their genesis and geochemistry, with deep neutral chloride waters usually containing higher As concentrations than shallow acidic sulfate waters (Wang et al., 2018). Geothermal waters in eastern Slovakia are of the sodium-chloride type with arsenic concentrations up to 36.7 mg/L (Vranovská et al., 2015).

Arsenic in deep geothermal waters can be released to surface waters when there is discharge of hot springs or geothermal wastewater, or via natural mixing of geothermal waters with local shallow groundwaters (Wang et al., 2018). Arsenic is mobilized from deep geothermal systems at low temperatures (150-250 °C) from arsenic bearing pyrite while at high temperature (>250°C), arsenic is mobilized from arsenopyrite (Bundschuh et al.,2016). High arsenic concentrations (Table 2) up to 162,000 µg/L were found in the volcanic geothermal systems of Los Humeros in Mexico (Bundschuh et al.,2016). Arsenic concentrations in geothermal waters from igneous rocks was higher than from sedimentary rocks (Maity et al., 2016). In the Bolivian Altiplano, arsenic concentrations in in cold water samples from shallow aquifers are higher than those in thermal springs (Munoz et al., 2015).

CONCLUSION

Geothermal energy driven by geothermal waters is a form of renewable energy which is fast gaining acceptance as an alternative to fossil fuel. However, sometimes geothermal water is used for urban water supply or for agricultural purposes in regions with water scarcity. In such situations, the quality of the geothermal water provided to the public is important to protect public health. Several studies have reported high arsenic concentrations above acceptable limits in geothermal waters which could have adverse effects on consumers if used for public water supply. Geothermal waters intended for use as urban water supply should be properly screened for arsenic contamination before use. Geothermal waters with arsenic concentrations higher than allowable limits should not be used for public water supply or for agricultural purposes.

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