



Hydrogeochemical and isotopic evaluation of arsenic contaminated waters in an argillic alteration zone



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ABSTRACT

Mineralization of sulfidic minerals including realgar, orpiment and pyrite occurs in argillic alteration zones in northeastern Iran, which affects water quality and health in these semi-arid localities. Geochemical source of ions in surface and ground waters was examined to evaluate potential effects of sulfidic mineralization on water quality. The surface and groundwater samples were analyzed to determine the major ions (Na^+ , Ca^{2+} , Mg^{2+} , K^+ , HCO_3^- , SO_4^{2-} , Cl^-) and trace elements (such as As, B, Br). Water-rock interactions and evaporation defined as the key phenomena on groundwater chemistry using Gibbs diagram. Concentrations of arsenic (As) varied from 16 to 606 $\mu\text{g/L}$, which was higher than the WHO (2011). Calculated ionic ratios revealed that anions and cations in surface and ground waters originated from partial leaching through ion exchange in alteration zones. We postulate that the primary source of As resulted from oxidation of As-bearing sulfide minerals such as orpiment, realgar and arsenopyrite in argillic-pyrite alteration zone. High pH (>8) could provide the alkalinity to increase sulfide oxidation and release As into the water. The stable isotope data ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) indicated the origin of the waters, which is mainly meteoric precipitation with partial effects from evaporation processes and exchanging ions with surrounding rocks. This confirms the notion that the source of all analyzed ions including the toxic As is geogenic. Hydrogeochemical process, which affected the water chemistry and thus environmental public health are likely to be water-rock interactions and evaporation.

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1. Introduction

Widespread arsenic (As) accumulation, mobility in sediments, water and air (dust) is a critical environmental public health issue, worldwide (Ravenscroft et al., 2009; Rodríguez-Lado et al., 2013; Taheri et al., 2016a). As a result of geological phenomena and environmental changes, high levels of geochemically dispersed As can be found in feed, foods, water, soil and air in both organic and inorganic forms (Hall et al., 2006; Fillol et al., 2010) reaching toxic levels to surrounding human and animal populations and adversely affecting biochemical processes of cells and organs. International Agency for Research on Cancer (IARC) has classified As as a Class I carcinogen (IARC, 2004). Owing to its carcinogenic properties, the maximum allowable concentration (MAC) value of As in drinking water is 10 $\mu\text{g/L}$ (Tamasi and Cini, 2004). The Environmental Protection Agency (EPA) and the National Research Council

(NRC) stated that chronic effects on humans may be caused by prolonged consumption of water with a concentration of As as low as 5 $\mu\text{g/L}$ (EPA) or even 3 $\mu\text{g/L}$ (NRC) (Hogue, 2001a, 2001b; Tamasi and Cini, 2004).

Water-rock interaction is one of the main factors controlling geochemical characteristics of the water. Indeed, one of the crucial areas of research in chemical geology is groundwater chemistry (Hounslow, 1995); application of groundwater hydrochemistry and how rocks/sediments physicochemically interacts with the water would multidisciplinary broaden our knowledge about environmental and public health. Although evaporation-based concentration and precipitation-related dilution could change the chemical compositions of groundwaters, rock/sediment-water interaction is nonetheless the major process because solid phases (inorganic and organic matter) are the primary sources and sinks of dissolved constituents in groundwater. The chemical reactions that are responsible for the water-rock interaction vary spatially and temporally, depending on the chemistry of the initial waters and dynamics of geological formations and residence time (Sarkar

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et al., 2007). The resulting concentrations of major ions of groundwater can be used to identify the intensity of water-rock interactions and chemical reactions.

Distinctive chemical composition of ground waters has been due to association with hydrothermal systems within fault zones, where interaction of hydrothermal fluids with the wall rocks and atmospheric water and subsurface brine occurs (Aiuppa et al., 2006). Chemical characteristics of spring water and hydrothermal alteration zones have been studied to evaluate their relationship. For example, oxidation of sulfide boosts SO_4^{2-} level, thus leading to precipitation of sulfate minerals such as gypsum (Karakaya et al., 2007). Hydrothermal activity and volcanic systems are typically associated with As release to the groundwater (Nriagu, 1989; Aiuppa et al., 2006). In addition to naturally occurring As, oxidation of arsenides and arsenosulfides such as orpiment (As_2S_3), realgar (As_4S_4) and arsenopyrite (AsFeS) in mine tailings are related anthropogenic As to the ground and surface waters.

Considering the broad hazardous effects of As on the natural environment and animal/public health (Argos et al., 2006; Banerjee et al., 2009; Taheri et al., 2016a), geochemical studies related to geogenic As contamination in vital water resources from this agro-ecologically important arid northeastern Iran, is warranted. We therefore intend to evaluate the geochemical source of As with other related major ions, and saturation parameters affecting the quality of surface and ground water resources in the highly mineralized region of Chelpu-Koohsorkh in northeastern Iran to rediscover a new window to understanding the geochemical mechanisms and functional consequences of As in inducing environmental health risks in As-exposed inhabitants.

2. Location and geological setting

Chelpu is located along the east longitude of $58^\circ 29' 44''$ to $58^\circ 36' 07''$ and north latitude of $35^\circ 33' 12''$ to $35^\circ 37' 77''$ (Fig. 1). This mountainous area demonstrates a semi-arid climate. Its dominant lithology consists of shale, limestone, sandstone, conglomerate and evaporites (gypsum and halite), from the Paleogene age.

There are numerous tectonic forms of anticline, syncline, and folded beds and in particular several faults in Chelpu. East-west trending strike slip "Doruneh fault" is the main fault accompanied by N-W to S-E oblique-slip and normal faulting system in the region (Aghanabati, 2004). Resulting fractures were filled by hydrothermal solutions leading to hydrothermal mineralization along the fractures. Epithermal mineralization of As, Sb and Au elements at temperatures $< 300^\circ\text{C}$ has been reported in stoke framework of veins in this area (Mazloumi Bajestani, 1992). Jarosite [$\text{KFe}(\text{SO}_4)_2\text{OH}_6$] and alunite [$\text{KAl}_3(\text{SO}_4)_3\text{OH}_6$] minerals together with montmorillonite and kaolinite (clay minerals) are hydrothermal alteration-minerals, present in this location that indicate hydrothermal activities in this region. Mineralization of sulfides such as realgar, orpiment, arsenopyrite, pyrite, and stibnite are accompanied with calcite and gypsum within the alteration zones, also known as argillic-pyrite alteration zones (Mazloumi Bajestani, 1992; Karakaya et al., 2007). In fact, the As-bearing sulfides (orpiment, realgar, pyrite and arsenopyrite) were hosted in the sedimentary rocks of shale, marly limestone, sandstone, conglomerate (Mazloumi Bajestani, 1992).

Stages of mineralization in the host rock are as following: 1) creation of fractures and hydrothermal fluid ascent through fractures, 2) stibnite

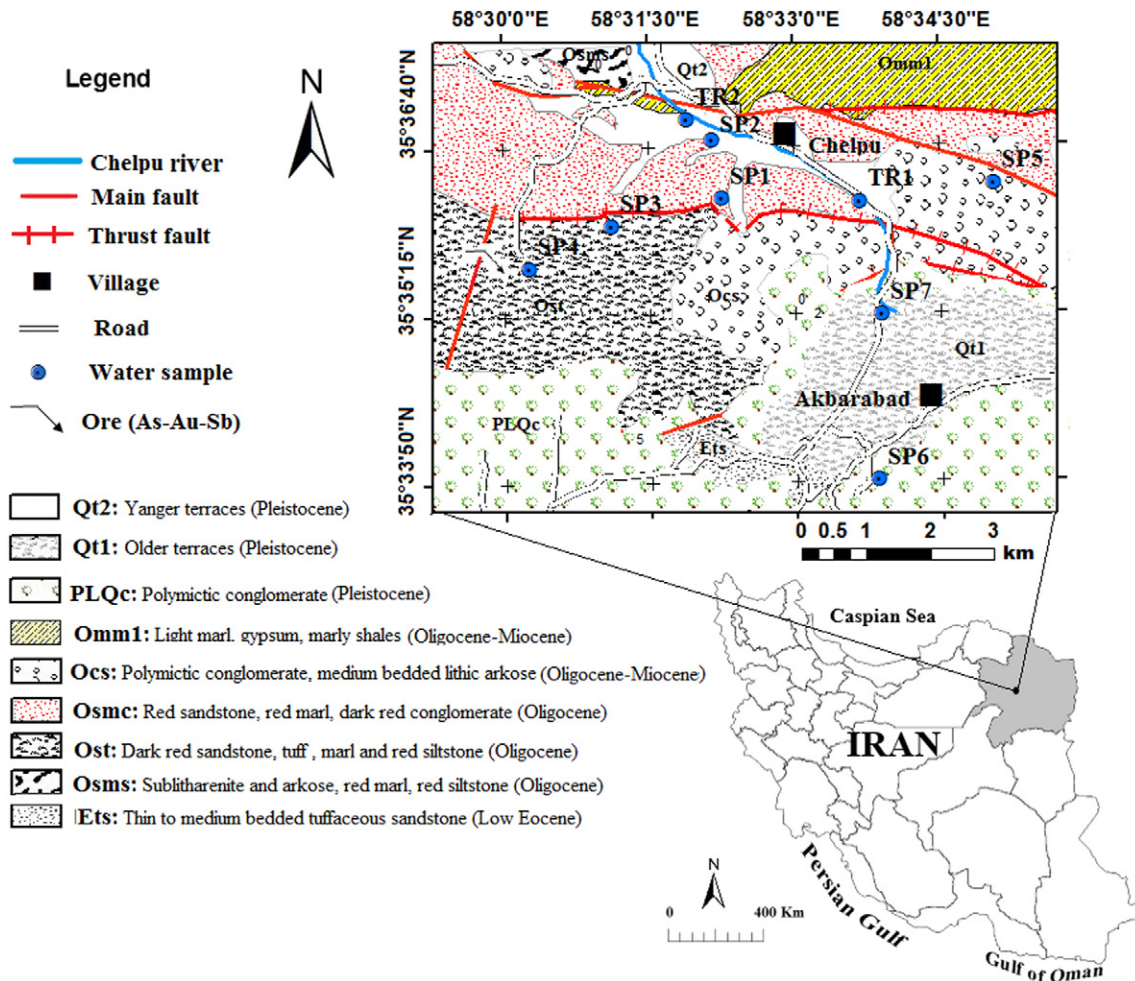


Fig. 1. Simplified Geological Map of Chelpu area (with slight modifications in drawing of 1:100,000 Shamkan and Kadkan map, Persian report of Geological Survey of Iran, N. Naderi Mighan, 1999a,b, #7661).

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