



Influence of tectonics, sedimentation and aqueous flow cycles on the origin of global groundwater arsenic: Paradigms from three continents



Abhijit Mukherjee^{a,*}, Swati Verma^a, Saibal Gupta^a, Kevin R. Henke^b, Prosun Bhattacharya^c

^a Department of Geology and Geophysics, Indian Institute of Technology Kharagpur, Kharagpur, WB 721302, India

^b Center for Applied Energy Research, University of Kentucky, Lexington, KY 40511-8479, USA

^c KTH-International Groundwater Arsenic Research Group, Division of Land and Water Resources Engineering, Department of Sustainable Development, Environmental Sciences and Engineering, Royal Institute of Technology (KTH), SE-100 44 Stockholm, Sweden

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SUMMARY

Arsenic (As) is a trace element in the Earth's crust. However, its presence in elevated concentrations in groundwaters of major aquifers around the world raises concern about its primary source(s). A close look at the global distribution of known As enriched areas reveals an intriguing systematic pattern, where most of the As enriched aquifers are parts of large sedimentary basins adjoining major orogenic belts, suggesting the existence of a large-scale geological process. Many of these sedimentary basins may be tectonically regarded as foreland basins that developed by lithospheric flexure at the time of mountain building processes (orogenesis) along convergent plate boundaries. Arsenic enrichment in the groundwater of these foreland basin aquifers may be ultimately sourced to crustal evolution processes related to plate tectonics, along with transportation of As-enriched magmatic rocks from the depth to surficial deposits, which subsequently release or mobilize the As to groundwater under conducive surficial biogeochemical processes. Circulating As-laden hydrothermal fluids may also be derived from magmas and form part of the discharge in the surficial hot springs in arc environments. These hydrothermal-genic deposits in orogenic belts may also act as the primary provenance for the As-laden sediments that are transported into foreland basins by wind, glacial erosion, and/or streams. Ultimately, the As-laden foreland sediments serve as modern-day aquifers, where the sediments release As into groundwater by water–rock interaction during various biogeochemical processes under conducive hydrogeochemical conditions. The significance of this hypothesis is that it proposes the existence of a common primary source for globally dispersed geogenic As-enriched aquifers, that have been so far mostly studied as individual occurrences. The proposed hypothesis can explain the widespread presence of As in areas as diverse as the Indus–Ganges–Brahmaputra basin (Himalayan orogen), the Chaco–Pampean basin (Andean orogen), Rocky mountain basin (Western Cordilleran orogen), New England and northeastern USA (the Appalachian orogen).

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1. Introduction

In general, arsenic (As) is present in trace concentrations (mean: ~1.8 mg/kg [Matschullat, 2000, Bhattacharya et al., 2002]) in the Earth's continental crust (e.g. Bhattacharya et al., 2002; Nordstrom, 2002; Smedley and Kinniburgh, 2002). However, hydrological systems, specifically groundwater systems, are often enriched in As (>0.01 mg/L). Over the past few decades, more than twenty geological provinces across the globe have been identified as having elevated, and sometimes toxic, concentrations of dissolved As in groundwater. The chemistry of groundwater is largely controlled by the composition of the recharging water, water–aquifer matrix interactions (e.g. cation exchange, mineral dissolution),

availability of reactive organic matter and groundwater residence time within the aquifer (e.g. Edmunds et al., 1981; Stallard and Edmond, 1983; Chapelle, 1993; Drever, 1997; Faure, 1998; Mukherjee et al., 2009, 2012). Accordingly, elevated concentrations of chalcophile As in contaminated aquifers may result from the dissolution of primary or secondary sulfidic minerals (such as pyrite), or desorption from secondary concentrations in hydrous metal oxides (Nordstrom, 2002), that are common mineral constituents of the aquifer matrix in many large, unconsolidated sedimentary systems. Some of the potential As source materials for water systems include mineralized areas (mostly sulfide), volcanogenic deposits and sediments, geothermal springs, organic-rich shales, authigenic sulfides in sediments and recent metal oxide/hydroxide-rich sediments in aquifers with low hydraulic gradients (Bhattacharya et al., 2004; Scanlon et al., 2009). However, it has been observed that As-rich groundwater are often separated from their sources, and it has

* Corresponding author. Tel.: +91 3222 283396.

E-mail address: amukh2@gmail.com (A. Mukherjee).

been justifiably noted that As mobilization mechanisms might be a more critical issue than the source (e.g. Smedley and Kinniburgh, 2002; Nordstrom, 2002; Mukherjee and Fryar, 2008; Scanlon et al., 2009; Ravenscroft et al., 2009). Hence, while there is great emphasis on uncovering the hydrogeochemistry that controls mobilization processes in contaminated areas, studies delineating the source of the As have remained uncommon. Saunders et al. (2005) commented that although there is a consensus that mobilization processes lead to As enrichment, the ultimate source of As is not well understood. It is obvious that even the trace or normal concentrations of As in solid phase aquifer matrices (Korte and Fernando, 1991) is sufficient to produce toxic As concentrations in groundwater under conducive environments (Stüben et al., 2003; Guillot and Charlet, 2007). However, it has been also repeatedly observed that certain portions of the geosphere are more enriched in As than others, and these enriched portions may, in part, the primary source for groundwater As.

Van Geen (2011) suggested that the two most important aspects of understanding the causes of high As groundwater is to identify whether the elevated As is reactively mobilized in the aquifer *in situ*, or if originally low-As areas have been contaminated by advectively transported As along flow paths from up-stream enriched sources (*ex situ*). Van Geen (2011) hypothesized that the spatial distribution of natural As in aquifers is largely controlled by mobilization processes with the possibility of local geologic influences, where the hydrochemical evolution leading to As enrichment in groundwater might take only a few decades to thousands of years. The importance of *ex situ* sources, in addition to the *in situ* mobilization mechanisms for As have been recognized in most previous global reviews (e.g. Smedley and Kinniburgh, 2002; Nordstrom, 2002; Mukherjee et al., 2008; Ravenscroft et al., 2009). Smedley and Kinniburgh (2002) noted that typical environments containing high As groundwater include (1) low temperature (including mining and non-mining) and (2) high temperature (hydrothermal) settings. High As groundwater in low temperature, non-mining areas are generally characteristic of young, sedimentary aquifers (Bhattacharya et al., 2004; Scanlon et al., 2009). This observation suggests that the cause of As in groundwater might not be linked to just “local” geology but may be a reflection of larger, regional-scale geodynamic processes. Ravenscroft et al. (2009) observed that many of the As-enriched areas are located in the proximity of young fold mountain belts, but did not elaborate on the relationship. Saunders et al. (2005) proposed that As-enriched groundwater is a product of interactions between tectonic, geochemical and biological processes. The geochemical processes are linked to chemical weathering of a variety of bedrock types, as noted by analyses of surficial material in major parts of continental crust (e.g. Ahmed et al., 2004; Grosz et al., 2004; Saunders et al., 2005; Mukherjee et al., 2012), which re-emphasizes the existence of some regional-scale geochemical process. In this context, a major debate concerns the source of the As-enriched aquifers of the major fluvial systems of Southeast Asia (e.g. Ganges–Brahmaputra, Indus, Mekong, Red, Irrawady, etc.), for many of which the Himalayas act as the sediment provenance and headwater (Fendorf et al., 2010). Notwithstanding these observations, only a few studies or rather hypotheses, have been proposed yet, to elucidate the Southeast Asian scenario (e.g. Stanger, 2005; Hattori and Guillot, 2003; Guillot and Charlet, 2007). Knowledge of the cause of As-enrichment in most other parts of the world is very limited.

Of the known global-scale, near-surface geological processes, tectonics associated with crustal plate movements are most important. Zheng (2006) and Nordstrom (2009) alluded to a link between As enrichment and crustal evolution. In spite of the heterogeneity of As concentrations based on lithotype and geography (e.g. Mukherjee et al., 2011), the known natural As enriched groundwater localities of the world (following Smedley and Kinniburgh, 2002;

Mukherjee et al., 2008; Ravenscroft et al., 2009) show a striking systematic relationship when plotted on the global tectonic map. Most groundwater As enriched areas exist in or near the sedimentary basins adjacent to orogenic belts (Fig. 1). This observation signifies a large-scale cause-effect relationship between global orogenesis and the concentration of arsenic in groundwater. The objective of this study is therefore to establish a hypothesis that identifies a common primary source-aggradation mechanism for the formation of different globally dispersed geogenic As-enriched aquifers that have been so far mostly studied and explained in terms of individual occurrences.

In geodynamics, the term “orogenesis” refers to the formation of long, mountain belts called orogenic belts or orogens. In plate tectonic terms, orogenesis is a manifestation of the convergence of tectonic plates, resulting in subduction or obduction of lithospheric plates. Present-day or examples include the Alpine–Himalayan orogen (Alps and Himalayas) that stretches from western Europe to eastern China, through the Indian subcontinent, the Cordilleran orogens along western North America, the Andean orogen of western South America, and the Lachlan orogen of southeastern Australia, among others. Ancient equivalents are the Appalachian of eastern USA, and the Caledonian orogen of northern Europe and eastern Greenland. Associated with these orogenic belts are extensive foreland basins, which are depressions that develop during the mountain building process by lithospheric flexure (DeCelles and Giles, 1996). These foreland basins act as sinks for the sediments that are eroded from the rapidly rising, adjacent orogens. The basins are characterized by thick sedimentary successions, which thin away from the orogenic belts. Examples of such foreland basins include the Indus–Ganges–Brahmaputra, Northern Tarim, Southern Junggar basins of Asia, the Northern Alpine, Po, Ebro, Guadalquivir, Aquitaine basins of Europe, the Appalachian and Western Canada Sedimentary basins of North America. Based on the structures (DeCelles and Giles, 1996), foreland basins may be classified as peripheral (where the basins occur on the plate that is subducted i.e. the outer arc of the orogen, e.g. the Indus–Ganges–Brahmaputra basin) and retroarc (where the basins occur on the plate, which overrides during the tectonic plate convergence, and are situated behind the magmatic arcs, e.g. the basins of the Andean or Western Cordilleran orogen). We hypothesize that the primary source of the As in groundwater is *ex situ* to the affected aquifers and globally, the groundwater As in the aquifers of the foreland basins might be derived from tectonic cycles, sedimentation and regional-scale water flow, and mobilized on availability of conducive surface geochemical and biogeochemical environments as illustrated, in this article, by the examples from three continents.

2. Hypothesis for primary source of arsenic in Earth's surface

2.1. Provenance

Arsenic has a tendency to be enriched in chalcophile mineralized zones in the Earth's crust. However, on an average, the continental crust has a solid phase As concentration of 1.0–1.8 mg/kg (Krauskopf and Bird, 1995; Wedepohl, 1995; Faure, 1998; Henke, 2009). Of these, the upper crust has been studied by many workers, and is in general As-enriched with average concentrations varying from 2 mg/kg (Wedepohl, 1995) to 5 mg/kg (Sims et al., 1990), in comparison to the less studied, As-depleted lower crust (1.3 mg/kg; Wedepohl, 1995). The granites and granodiorites of the upper crust have average As concentrations of 3 mg/kg (Matschullat, 2000). In contrast, basaltic rocks in the upper crust have a relatively lower, average As concentration of 2.2 mg/kg (Faure, 1998). Mid-oceanic ridge basalts have an average As content of ~1 mg/kg

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