



# A Critical Evaluation of the Role of Geotectonics in Groundwater Arsenic Contamination

Mohammad Ayaz Alam, Abhijit Mukherjee, and Prosun Bhattacharya

## Abstract

It has been observed globally that primary groundwater arsenic (As) is derived from modern and ancient magmatic arcs at continental convergent margins of some of the most prominent orogenic systems worldwide (e.g., Andes, Himalaya). It ends up in arc-derived sediments in the adjacent foreland basin through rapid and continuous erosion of exhuming arc sequences, where groundwater could be contaminated through secondary As in the aquifer matrix, mostly allochthonous arc-derived sediments. Apart from the origin and development of magmatic arc, geotectonic control for other primary As sources is also evident in the form of (i) global distribution of geothermal and volcanic centers, where As-enriched hydrothermal and magmatic fluids contaminate surface and groundwater, (ii) ore (especially metal sulfide) deposits through natural leaching that releases As to surrounding environments, which is accelerated by metal mining and processing activities, and (iii) coal deposits and hydrocarbon reservoirs, together with the exploitation processes of both these fuel resources, are other geogenic sources of As contamination that is

accelerated by the extraction and refining processes and subsequent usage. Whereas the aforementioned solid and liquid phase As inputs to groundwater are attributed to the lithology and geothermal and volcanic fluids, respectively, As speciation can also be associated with the basin's structural settings driven by geotectonics. Basin morphology, prolongation of faults, especially division of the underlying basement into blocks can produce discrete compartmentalized hydrodynamic environments, resulting in spatial variations in hydrochemical composition, including As speciation.

## Keywords

Arsenic • Environment • Contamination • Groundwater • Geotectonics • Speciation • Arc magmatism • Geothermal • Volcanism • Metallogeny • Orogenesis • Foreland basin • Basement structure

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## 1 Introduction

Although arsenic (As) content of crustal rocks varies widely, for most known and quite extensive instances of groundwater As contamination, its sources are geogenic (Smedley and Kinniburgh 2002; Bhattacharya et al. 2002, 2017; Nriagu et al. 2007; Coomar et al. 2019). Arsenic is the most ubiquitous toxic element present in all sorts of geological environments that gets transported by all kinds of media. Tracing all its origin and pathways for its human exposure has been a challenging task. However, this challenge has been met through multidisciplinary global collaborative and team efforts by deciphering As sources and pathways. Arsenic can get disseminated in air and carried by wind (e.g., volcanic ash in Argentina and Chile, Bhattacharya et al. 2006a, b, Quintanilla et al. 2009; Bundschuh et al. 2004, 2010, 2012; Nicolli et al. 2012; Morales-Simfors et al. 2020; Aullón Alcaine et al. 2020; Mariño et al. 2020), dissolved

and transported by water (e.g., groundwater in Argentina, Bangladesh, Bolivia, Chile, India, Peru, Tanzania, Bhattacharya et al. 1997, 2001; Biswas et al. 2014a, b, c; Chakraborti et al. 2015; Bhowmick et al. 2018; Tapia et al. 2019a, b, Chakraborty et al. 2015, 2020; Mukherjee et al. 2021; Quino Lima et al. 2020, Quino Lima 2021a, b, Ijumulana et al. 2020), or stay in solid form in As-bearing minerals (e.g., mineralized zones, coupled with the residual products of mining and metallurgical processes, e.g., Latin America and the Caribbean, Quintanilla et al. 2009; Ramos et al. 2012, 2014; Bundschuh et al. 2012, 2020).

The ubiquitous nature of As in the environment can be understood simply by considering that hypothetical leaching of its average crustal concentration (1.8 mg/kg, Henke 2009) into groundwater is sufficient to cause toxic levels of As concentration (0.01 mg/L, WHO 2017). Moreover, surface sediments and soils in various parts of the world contain higher and more variable concentrations (ranging from 2 to 20 mg/kg on average) than the average crustal concentration of 1.8 mg/kg (Taylor and McLennan 1985; Wedepohl 1995; Mandal and Suzuki 2002; Bhattacharya et al. 2002; Henke 2009; Mukherjee et al. 2009a, b, and references therein). The average and maximum As concentration in 3,024 marine sediment samples from coastal Japan was determined to be 14 and ~580 mg/kg, respectively, of which As concentration in 65% samples was <10 mg/kg, while in 87.5% it was <20 mg/kg (Masuda 2018). On the other hand, Ohta et al. (2010) attributed the sediments with high As concentrations in Japan's inland areas to coal and sulfide mines. Consequently, even a trace amount of As in solid-phase aquifer matrices is sufficient for groundwater under conducive environments (Korte and Fernando 1991; Stüben et al. 2003; Sracek et al. 2004; Guillot and Charlet 2007; Henke 2009). All these imply that any lithological unit in the crust can be considered a potential As source (Matschullat 2000; Bhattacharya et al. 2002, 2007; Nordstrom 2002; Smedley and Kinniburgh 2002; Saunders et al. 2005). Nonetheless, considering "average value" being a theoretical concept and also, as a matter of fact, not all aquifers around the world are As-contaminated, as only certain segments of the lithosphere are enriched in As. Delineation and segmentation of such enriched portions are the basis for discussing the role of geotectonics in As contamination of aquifers in this chapter, based on some insightful works and representative examples from across the world (e.g., Mukherjee et al. 2007, 2008, 2009a, b; Mukherjee et al. 2014, 2019; Barringer and Reilly 2013; Giménez-Forcada and Smedley 2014; Masuda 2018; Morales-Simfors et al. 2020; Bundschuh et al. 2020).

## 2 Some Considerations and Constraints

Arsenic, a metalloid, being siderophile and chalcophile element, behaves as a metal and combines with sulfur; however, it shows volatile behavior in molten rock, similar to the light rare earth elements (Sims et al. 1990; Bhattacharya et al. 2002; Nriagu et al. 2007). The partition coefficient of As between sulfide and silicate melts varies as a function of oxygen fugacity,  $f_{O_2}$  (Li and Audétat 2012, 2015). Accordingly, at a specific temperature and FeO content, As fraction in silicate and monosulfide melts increases with  $f_{O_2}$  (Li and Audétat 2012, 2015). Although silicate and carbonate mineral phases do not host As as a major element, i.e., it does not appear in their formula (Majzlan et al. 2014), at low temperature, high  $f_{O_2}$  and low sulfur fugacity  $f_{S_2}$ , silicates and carbonates can incorporate As, e.g., filatovite  $K(Al,Zn)_2(As,Si)_2O_8$ , a variety of feldspar first reported at Tolbachik volcano, Kamchatka, Russia (Vergasova et al. 2004). Charnock et al. (2007) found As incorporated into garnet in the overgrowth of a hydrothermal vein. Pascua et al. (2005) reported As-enriched smectite in hydrothermal precipitates at a geothermal plant in Akita, Japan. Moreover, Ryan et al. (2011) found fractured aquifers in ultramafic rocks contaminated with As derived from serpentine and magnesite through chemical weathering.

Arsenic sources that can potentially contaminate the aquifers, i.e., leading the As concentration to toxic levels (0.01 mg/kg, WHO 2017), may develop through complex interactions between tectonic, geochemical, and biological processes (e.g., Saunders et al. 2005; Zheng 2006; Nordstrom 2009; Barringer and Reilly 2013; Mukherjee et al. 2014, 2019; Masoodi and Rahimzadeh 2018; Masuda 2018; Polya et al. 2019). Although there is no controversy about As mobilization from the aquifer matrix leading to As enrichment of groundwater and there are plenty of works on it (e.g., Smedley and Kinniburgh 2002; Saunders et al. 2005 and references therein), the primary source of As is often controversial and studied by few researchers in detail (e.g., Mukherjee et al. 2011; Raychowdhury et al. 2014). Bundschuh et al. (2021) have identified different sources and pathways for As exposure for Latin America, namely, (i) volcanism and geothermalism, taking into account (a) volcanic rocks, fluids (e.g., gases), and ash, including large-scale transport of the latter through different mechanisms (b) geothermal fluids and their exploitation, (ii) natural lixiviation and accelerated mobilization from (mostly sulfidic) metal ore deposits by mining and related activities, (iii) coal deposits and their exploitation, (iv) hydrocarbon reservoirs and co-produced water during exploitation, (v) solute and sediment transport through rivers to the sea,

(vi) atmospheric As (dust and aerosol), and (vii) As exposure through geophagy and involuntary ingestion. Of these, authors identified the two most important and well-recognized sources and mechanisms for As release into environments affecting the Latin American population: (i) volcanism and geothermalism, and (ii) the strongly accelerated release from geogenic sources by mining and related activities. This is in line with the findings of Smedley and Kinniburgh (2002) that typical environments containing highly As-contaminated groundwater include (i) low temperature (including mining and non-mining) and (ii) high temperature (hydrothermal) settings.

Since As-contaminated groundwater is often distal from the (primary) As source(s), it has been argued continuously that understanding As mobilization is more critical than deciphering the (primary) source (e.g., Nordstrom 2002; Smedley and Kinniburgh 2002; Mukherjee and Fryar 2008; Scanlon et al. 2009; Ravenscroft et al. 2009). Consequently, many published peer-reviewed works deal with the hydrogeochemical processes controlling As mobilization processes in the contaminated areas, which hugely outnumber the studies on primary or ultimate As source. Saunders et al. (2005) emphasized the need for the latter, putting forward a case for a better understanding of the ultimate As source, considering that there is a consensus about mobilization processes leading to As enrichment within the aquifers (Saunders et al. 2005). van Geen (2011), on the other hand, put this view in another term. He emphasized the importance of understanding the etiological aspects of As enrichment in groundwater and the mobilization in aquifers and its control through in-situ reactive mobilization within the aquifer or transported advectively along flow paths from upstream enriched sources, i.e., ex situ, to areas with low As initially. van Geen (2011) proposed that mobilization processes, viz. water chemistry, with possible influence of local geology, lithology in particular, control the spatial distribution of As in aquifers. The duration of such hydrochemical processes leading to As enrichment of groundwater could be from few decades to thousands of years (van Geen 2011). Several workers have recognized the importance of ex situ sources, in addition to the in situ mobilization processes (viz. leaching) for As enrichment of groundwater, e.g., Smedley and Kinniburgh (2002), Nordstrom (2002), Mukherjee et al. (2009a, b), Ravenscroft et al. (2009). Accordingly, high As in young sedimentary aquifers should not be attributed only to local geology, as the time required for in situ hydrochemical processes might be more than the time elapsed since the deposition of those sediments and diagenesis, instead to larger, regional-scale geodynamic processes (Bhattacharya et al. 2004; Scanlon et al. 2009; Mukherjee et al. 2014, 2019). In this regard, although Ravenscroft et al. (2009) did highlight the proximity of young fold mountain belts to many As-enriched areas, they did not elaborate

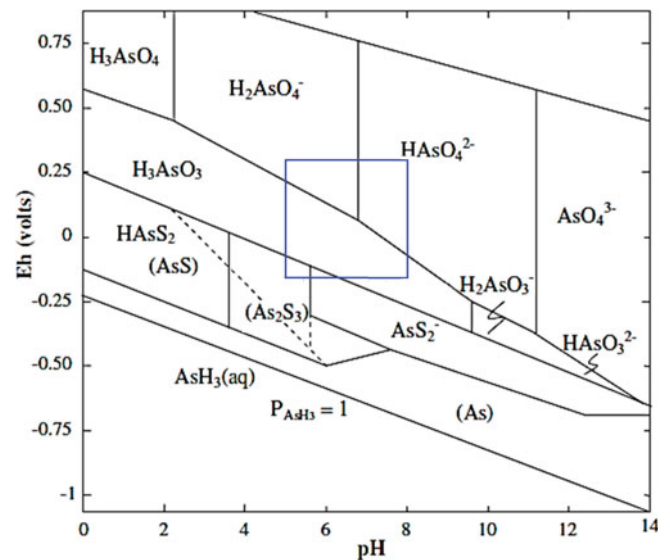
further on the relationship between the two. Saunders et al. (2005), on the other hand, proposed that As-contaminated groundwater is a product of interactions between tectonic, geochemical, and biological processes. Zheng (2006) and Nordstrom (2009) suggested a link between As enrichment and crustal evolution. Moreover, regional-scale geochemical processes lead to chemical weathering of a variety of As-bearing rocks, as noted by analyses of surficial material (viz. rocks, sediments, etc.) in different parts of the continental crust (e.g., Ahmed et al. 2004; Grosz et al. 2004; Saunders et al. 2005; Mukherjee et al. 2014, 2019). For example, As source in the contaminated aquifers of the major fluvial systems of the Indian subcontinent (e.g., Ganges–Brahmaputra, Indus, etc.) have been traced to the Himalayas, which acts as the sediment provenance, as well as the headwater for the rivers draining the basin (Stanger 2005; Hattori and Guillot 2003; Guillot and Charlet 2007; Fendorf et al. 2010; Mukherjee et al. 2014, 2019; Chakraborty et al. 2015, 2020).

Arc magmatism and related hydrothermal activity, together with high rates of erosion due to rapid uplift of the arc zones and transport of the eroded sediments to the downstream basins, contribute to As contamination of the aquifers through in situ leaching of As-bearing minerals present in arc derived sediments (Mukherjee et al. 2019). However, geotectonics play a significant role in arc magmatism and erosion rates of the arc zone lithologies (Hecht and Oguchi 2017; Masuda 2018), which implies the tectonic control on geodynamic processes responsible for the geologic distribution and dissemination of As.

Another vital consideration is As speciation, since not all its species are equally toxic to humans, animals, and plants. Moreover, its toxicity varies widely with its oxidation state. WHO (1981, 2001) has documented that inorganic species are generally more toxic than organic ones, and arsenite [As(III)] is 60 times more toxic than arsenate [As(V)], which is 70 times more toxic than methylated species, monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), which are consequently considered to be moderately toxic, whereas arsenobetaine (AsB) and arsenocholine (AsC) are considered to be nontoxic (Kumaresan and Riyazuddin 2001). Therefore, for an accurate environmental and human health risk assessment, identification and quantification of the As species are essential, especially the more toxic ones, rather than simply reporting total As concentrations, which has been the norm for many years. In this regard, Eh–pH diagrams (also known as Pourbaix diagrams, Pourbaix 1974) are conventionally used to represent and describe the speciation of As and identify significant geochemical processes (Welch and Stollenwerk 2002; Nicolli et al. 2012). Figure 1 shows the Eh–pH diagram of aqueous As species in water at 25 °C.

Arsenic may exist with oxidation states of  $-3$ ,  $0$ ,  $+3$ , and  $+5$ ; however, the oxidation states  $-3$  and  $0$  (native As) are

**Fig. 1** Eh–pH diagram illustrating the stability fields of the oxidized and reduced species of As in water at 25 °C. The blue rectangle represents the general pH and redox condition of groundwater systems. Modified after Herath et al. (2016)



rare in nature (Panagiotaras et al. 2012). Consequently, As chemistry primarily focuses on As(V) or As(III), depending on redox potential and pH of groundwater (Sracek et al. 2004). Both As(III) and As(V) may form protonated oxyanions in aqueous solutions (Panagiotaras et al. 2012). However, the extent of protonation depends mainly on the pH of the medium. Under oxidizing conditions, As(V) hydrolyzes to four possible species, depending on the pH range encountered in surface and groundwater systems. In oxygenated water systems, arsenic acid ( $\text{H}_3\text{AsO}_4$ ) is the predominant species only at extremely low pH (<2), whereas, As exists in the form of  $\text{H}_2\text{AsO}_4^-$  and  $\text{HAsO}_4^{2-}$  in a pH range of 2–11. In reducing environments and at low pH, arsenic acid converts into  $\text{H}_2\text{AsO}_3^-$ , and at pH > 12, it occurs in the form of  $\text{HAsO}_3^{2-}$  (Welch and Stollenwerk 2002; Panagiotaras et al. 2012; Gong et al. 2006). In the pH range of 4–10, As(V) species is negatively charged in water, whereas the predominant As(III) species is neutral because of redox potential and the presence of complexing ions such as sulfur, iron, and calcium and microbial activity (Katsyiannis and Zouboulis 2015). However, complete dissociation of As(V) is rare, because very few water systems reach a pH > 11.5 (Brookins 2012). In reducing environments, completely protonated arsenite species predominate over a wide range of pH without being ionized; hence, arsenite cannot adsorb As strongly as arsenate species, thus making the removal of the As species more difficult during remediation of As-contaminated water (Maity et al. 2011a). These authors found high As concentration in the geothermal fluids in southern Taiwan to be associated with low redox (Eh) conditions (Maity et al. 2011b).

Sulfide and arsenide minerals have low solubility in water and are insoluble in anoxic environments, e.g., the solubility coefficient (log K) of realgar ( $\text{AsS}$ ) and orpiment ( $\text{As}_2\text{S}_3$ ) is

–83.13 and –180.43, respectively, under standard temperature and pressure (STP) conditions, however, these minerals readily oxidize and then dissolve in oxic waters at STP (Boyle and Jonasson 1973). During chemical weathering of ore deposits and subsequent soil formation, As-bearing mineral phases change as follows: arsenopyrite and löllinite in the ore body become scorodite, pharmacosiderite, and arsenosiderite in the weathered rock, and finally goethite in the soil, latter As-bearing phases being more susceptible to getting dissolved and may contaminate surface and groundwater (Majzlan et al. 2014). Under neutral to alkaline conditions, arsenate ion substitutes for sulfate ion in the alteration minerals (Fernández-Martínez et al. 2008; Raychowdhury et al. 2014). Arsenite and arsenate minerals (e.g., scorodite) are the most likely sources of As contamination for their high solubility in aqueous solution, except under strongly acidic conditions (Masuda 2018). Due to secondary enrichment and their higher solubility, weathering products of the primary As-bearing rocks and minerals are more critical (and direct) sources of As contamination (Masuda 2018). For example, Savage et al. (2000) found that As concentration in soils, sediments, and weathered crust (>2000 mg/kg) from the fluvial basins in the Mother Lode District, California, have higher than nearby source rocks (<600 mg/kg).

In contaminated Cenozoic aquifers, As is primarily present in iron oxyhydroxides/oxides (usually goethite) through chemical weathering of iron-bearing minerals (Masuda 2018). Some studies examining iron oxyhydroxides/oxide precipitation and As incorporation into the aquifer sediments are as follows. According to Berg et al. (2007), in the Red River delta, the principal mechanism of As contamination of groundwater is the reduction of As-bearing iron oxyhydroxide minerals in the delta sediments, which are likely



chemical weathering products transported from an upstream source. Other mechanisms include in situ decomposition of As-bearing pyrite with seasonal changes of the groundwater level (e.g., Munshuganj, Bangladesh, Pollizzotto et al. 2005) and in situ chemical weathering of chlorite (e.g., Sonragaon, Bangladesh, Masuda et al. 2012, 2013). Both the Ganges–Brahmaputra delta examples imply that the source of As contamination in the aquifers is in situ dissolution of As from primary minerals rather than the dissolution of or desorption from As-bearing iron oxyhydroxides. Such dissolutions could be accelerated further through enhanced inductive recharge of surface water if there is overexploitation of the aquifers (Masuda 2018).

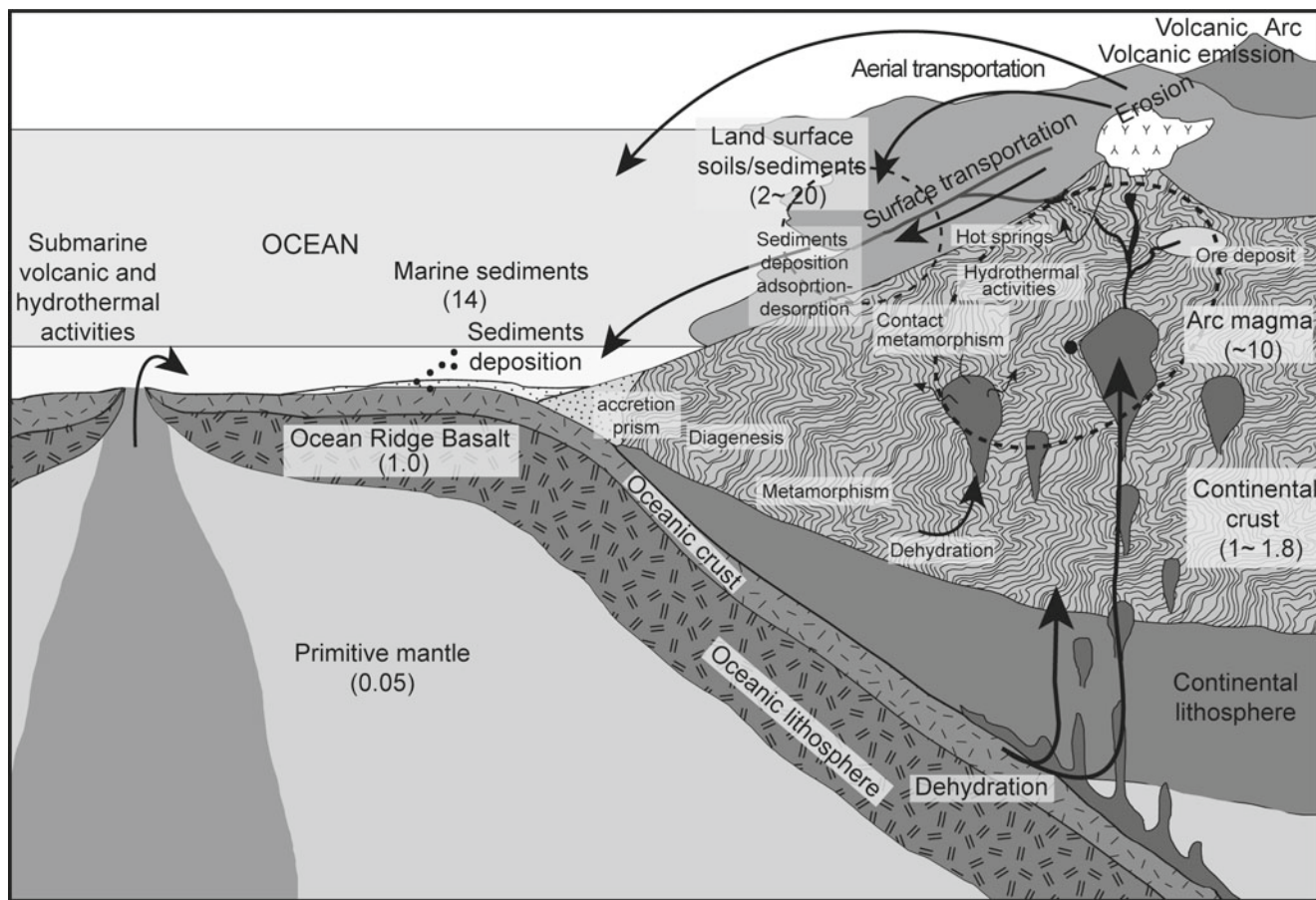
### 3 Geotectonic Arc Distribution and Recycling

The As concentration of Earth's primitive mantle is estimated to be 0.05 mg/kg (McDonough and Sun 1995). Hattori et al. (2002) found As highly concentrated in microscale sulfide minerals in mantle xenoliths from Lesotho, moreover, relatively higher As concentration in Iherzolite (>200 mg/kg As) than harzburgite (<14 mg/kg As). Besides, comparative analyses of mantle xenoliths of Ichinomegata, Japan by Hattori et al. (2002) showed that As is depleted in wedge mantle, where As concentrations of <50 mg/kg were found in microscale sulfide minerals.

Noll et al. (1996) showed enriching arc magmas in As (~10 mg/kg) by hydrothermal fluids derived from dehydrating subducting slab during metamorphism and confirmed the As release from subducting slab through laboratory experiments as well as analyses of the field samples. Through a similar experiment, You et al. (1996) demonstrated As enrichment of a hydrothermal solution at temperatures ~300 °C by leaching the sediments from the Nankai Trough (where the Philippine Sea plate is subducting beneath the Eurasian plate) with a synthetic NaCl–CaCl<sub>2</sub> solution. With the example of the relatively cool subduction zone associated with Catalina Schist on Santa Catalina Island of the Channel Islands, California, Bebout et al. (1999) proposed that devolatilization of As in fore-arc is low, so As is subducted at depth, resulting in As-enriched arc lavas in such subduction zones. On the other hand, Hattori et al. (2005) attributed the formation of As-bearing serpentine to alteration of the fore-arc mantle in the Himalayas, wherein As was derived from hydrothermal fluids releasing from the subducting slab, as observed in the aquifers of Ladakh Himalayas Indus-Tsangpo Suture Zone (Lone et al. 2020). This was corroborated by the documentation of an 85% (in some cases as much as 95%) decrease in the As concentration of arc lavas from fore-arc to back-arc

along the convergent margins (Noll et al. 1996). Fujiwara et al. (2011) reported As release from deep continental and island-arc crust during the low-temperature and high-pressure metamorphism (forming pelitic schists) in the rocks from the Sambagawa metamorphic belt, Japan. These authors also reported a decrease in As concentration from ~10 mg/kg to 0 (below detection limit) with the increasing temperature (from 300 to 450 °C). In striking contrast, Breuer and Pichler (2013) reported that As concentration in submarine hydrothermal fluids from the East Pacific Rise (up to 80.5 µg/L) and the Mid-Atlantic Ridge (up to 24 µg/L) one degree lesser than those from the back-arc basin (1386 µg/L) and island-arc (5850 µg/L). Sigfusson et al. (2011) found that As in hydrothermal solutions in basalts of Iceland are complexed with sulfur, i.e., As(OH)S<sub>2</sub><sup>2-</sup>, AsS<sub>3</sub>H<sup>2-</sup>, AsS<sub>3</sub><sup>3-</sup>, As(SH)<sub>4</sub><sup>-</sup>. The chemical species formed by covalent bonds between As and sulfur (S) precipitate from the hydrothermal solution as arsenide and sulfide minerals; consequently, many sulfide minerals, viz. pyrite, contain As (Masuda 2018). With decreasing temperature and increasing f<sub>O<sub>2</sub></sub> of the hydrothermal fluids, As–S complexes are decomposed and oxidized to arsenites, commonly found near the hot-spring water, where these fluids discharge on the surface (Masuda 2018).

High As concentration in hydrothermal systems than its average concentration in groundwater is reported globally (Maity et al. 2017). Arsenic-enriched hydrothermal discharges impact downstream surface and groundwater mixing, including related hydro(bio)geochemical processes (Maity et al. 2017). These processes are further influenced by respective changes in pressure, temperature, redox, and pH conditions (Mukherjee and Fryar 2008; Shimada et al. 2004). Such impacts of geothermal water on freshwater resources (surface and groundwater) lead to significant degradation, making affected resources unsuitable for drinking or irrigation without treatment (Maity et al. 2017). Hydrothermal discharges, even with similar geological/tectonic settings, may have very different As concentrations (Bundschuh and Maity 2015; López et al. 2012; Maity et al. 2011a, b; Maity et al. 2016, 2017). For example, significantly higher As concentrations (up to 75 mg/L) have been recorded for the hydrothermal discharges at the western margin of the Pacific Plate, e.g., El Tatio in northern Chile, Copper River and Yellowstone National Park in the USA, Los Humeros in Mexico (Ellis and Mahon 1967; Ball et al. 1998; Motyka et al. 1998; Gonzalez-Partida et al. 2001; Romero et al. 2003), as compared to those (up to 6.2 mg/L) at its eastern margin, e.g., Wairakei, Waiotapu, Ohaaki and Broadlands in New Zealand, Mt Apo in the Philippines (Ritchie 1961; Ellis and Mahon 1977; Webster 1990, 1999; Guo et al. 2007). A better understanding of As cycling in hydrothermal discharges



**Fig. 2** Redistribution of As during endogenic and exogenic geologic processes at the convergent plate boundary. Numbers in parenthesis indicate average As concentrations in the representative geologic

bodies (references within this section). Purple and blue colored letters indicate the releasing and fixing mechanism, respectively, of As with source to sink redistribution cycle. Modified after Masuda (2018)

will help better understand hydrothermal fluids' role in the geochemical evolution of global hydrological systems and the related environmental risk (Bundschuh et al. 2013; Maity et al. 2017) (Fig. 2).

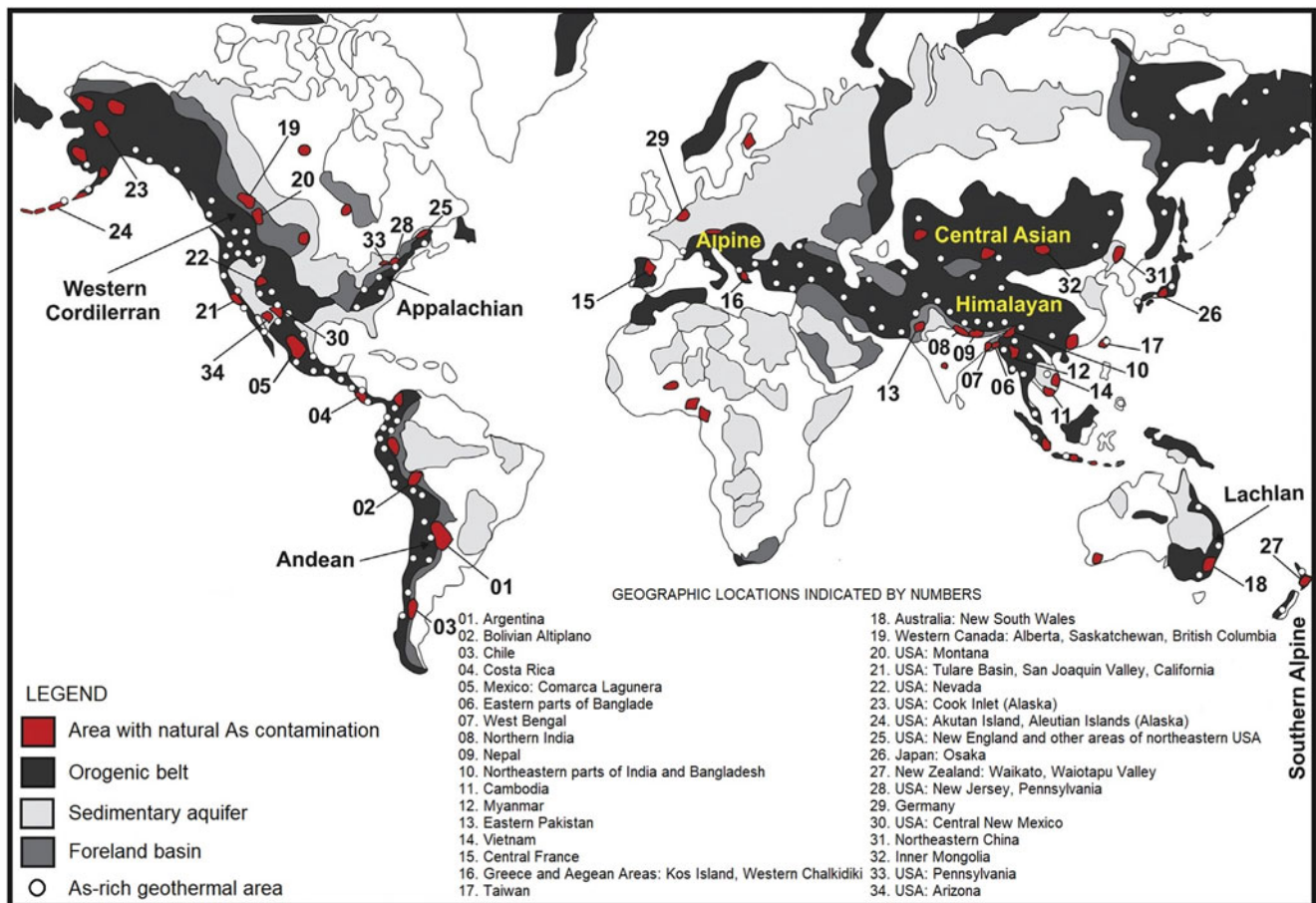
#### 4 Geotectonic Distribution and Disposition of As-Contaminated Aquifers

Despite intrinsic heterogeneity of As concentrations (primarily due to local geology) in more than 70 countries on six continents, worldwide distribution of naturally As-contaminated aquifers show their existence within foreland basins adjacent to the orogenic belts on the global tectonic map (Fig. 3; Smedley and Kinniburgh 2002; Mukherjee et al. 2014, 2019; Ravenscroft et al. 2009).

A large-scale source-sink relationship between orogenesis and As concentration in groundwater has motivated workers like Mukherjee et al. (2007, 2008, 2009a, b, 2011, 2012, 2014, 2019, 2021) to identify universal primary As source-aggradation mechanism(s) for contaminated aquifers

across the globe, instead of explaining individual cases of As contamination. With three examples, these authors corroborated that the primary As source in groundwater is ex situ. Also, As in affected foreland basin aquifers is derived through a series of geodynamic processes, viz. tectonic cycles, sedimentation, and regional-scale water flow. This As mobilizes with the availability of conducive surface geochemical and biogeochemical environments (Mukherjee et al. 2014, 2019). In this context, it is essential to consider modern and ancient orogenic belts and corresponding foreland basins. Present-day examples include (i) the Alpine-Himalayan orogen that stretches from Western Europe (the Alps), through the Indian subcontinent, to eastern China (the Himalayas), (ii) the Cordilleran orogens in western North America, (iii) the Andean orogen of western South America, and the (iv) Lachlan orogen of southeastern Australia, etc. Ancient equivalents are the Appalachian of the eastern USA and the Caledonian orogen of northern Europe and East Greenland (Mukherjee et al. 2014, 2019).

The aforementioned orogenic belts have associated foreland basins, which are extensive depressions developed



**Fig. 3** Global distribution of major As contaminated aquifers (groundwater resources data from BGR & UNESCO under World-wide Hydrogeological Mapping and Assessment Programme (WHYMAP) available at <http://www.whymap.org>, Margat 2008), orogenic belts (based on the global tectonic map available on the website of the USGS Earthquake Hazard Program, <http://earthquake.usgs.gov>), in addition to naturally As-enriched aquifers and geothermal fluids (based on Smedley and Kinniburgh 2002; Nriagu et al. 2007; Mukherjee et al. 2007, 2008, 2009a, b, 2011, 2012, 2014, 2019; Ravenscroft et al. 2009; Morales-Simfors et al. 2020). Modified after Mukherjee et al. (2019)

during the mountain building process through lithospheric flexure (DeCelles and Giles 1996). These foreland basins act as sinks for the sediments eroded from the rapidly exhuming adjacent orogens and typically have thick sedimentary successions, thinning away from the orogenic belts. Examples of such 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 their structures, DeCelles and Giles (1996) classified foreland basins as (i) peripheral, where the basins occur on the subducted plate, i.e., the outer arc of the orogen, e.g., the Indus–Ganges–Brahmaputra basin and (ii) retroarc, where the basins occur on the overriding plate during the convergence and are situated behind the magmatic arcs, e.g., the basins of the Andean or Western Cordilleran orogens.

Arsenic, being an incompatible element, is mainly concentrated in differentiated magmas; and as a result, volcanic rocks of intermediate (andesitic) to felsic (rhyolitic) composition have relatively higher As concentrations when compared to mafic (basaltic) and ultramafic rocks (Mukherjee et al. 2019), as shown by Welch et al. (1988, 2000) through a dataset of lava samples obtained from the western USA. This association of As with felsic rocks has been corroborated by Peters et al. (1999), Peters and Blum (2003), and Peters (2008), who attributed the As content in groundwater of Maine and New Hampshire, USA, to pegmatites (up to 60 mg/kg As), granites (46 mg/kg As) and granofels (39 mg/kg As), and also by Brown and Chute (2002), who attributed groundwater As to the weathering of pegmatite veins in Connecticut, USA. Moreover, in California, USA, As concentrations up to 48 mg/L have been reported in groundwater associated with acid volcanics, viz.

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tuffs and rhyolites, containing As-bearing sulfide minerals as the primary source of As (Welch et al. 1988, and references therein). On the other hand, it must be emphasized that although As content of mafic rocks is known to be relatively low, as pointed out by Welch et al. (1988, 2000), up to 327  $\mu\text{g/L}$  of As in groundwater has been attributed to fractured ultramafic rocks in Vermont, USA, by Ryan et al. (2011).

## 5 Arsenic Sources and Associated Geotectonic Settings

### 5.1 Metal (mostly Sulfidic) Ore Deposits

Ore deposits are one of the significant and widely recognized sources of As contamination, e.g., As released from marcasite, present in 5 Ma old hydrothermal ore deposits in New Zealand, causes groundwater contamination with  $>100 \mu\text{g/L}$  As (Craw et al. 2000). Although the distribution of geogenic As contamination around localized ore bodies is limited and defined mainly through geotectonic setting prevalent at the time of their formation, anthropogenic materials associated with ore mining and processing, viz. tailings and sludge, expand the area of contamination into the hydrosphere, thus affecting surface and groundwater, and pedosphere, i.e., soil horizons and near-surface ecosystems (Masuda 2018). Moreover, chemical weathering of the excavated ore bodies, exposed to exogenic forces of weathering and erosion, produce As-contaminated sediments, which might be the potential sources of As contamination of the environment and consequent human As exposure (Masuda 2018). Mining and metallurgical activities cause As contamination to spread to air, sediments, and streams, which might remain contaminated long after the cessation of these activities. For example, long after mining was ceased at Toroku mine, Miyazaki (Kyushu, Japan), 146 people were found to be suffering from arsenicosis due to inhalation of refining soot and drinking contaminated water at this site in 1990, where arsenite was mined from the 1820s to 1962 (Shimada 2009). On the other hand, in the absence of such activities, the contamination is restricted to a limited area (locality), close to the exposure or contact of ore bodies and alteration zones to water (surface or groundwater) or air.

Metal mining (mainly gold, copper, silver) in Latin America has been the key for As mobilization from the geologic materials and aggravating contamination of groundwater resources (McClintock et al. 2012; Tapia and Audry 2013; Tapia et al. 2012, 2018, 2019a, b, c, 2021). A summary table of the worldwide occurrence of As in groundwater by Nordstrom (2002) lists As bearing ores and other geologic sources of As, which include 24 As-bearing minerals that commonly occur in hydrothermal veins, ore

deposits, and rocks and dominate the environmental sources and conditions responsible for As mobilization to groundwater. According to the Committee on Medical and Biologic Effects of Environmental Pollutants of the National Research Council (NRC 1977), high concentrations of As are found in sulfide deposits, in native form or as alloys (4 minerals), arsenides (27 minerals), sulfides (13 minerals), sulfosalts (sulfides of As with lead, copper, silver and other metals, 65 minerals), and their oxidation products (2 oxides 11 arsenites 116 arsenates, 7 silicates). Altogether, As has been shown to occur in 568 different mineral species (Nordstrom 2002).

Most primary As minerals correspond to metal sulfides, which present As in solid solution with other metals, of which arsenopyrite is the most common (Ehrlich and Newman 2009). In contrast, secondary minerals tend to be less common and are related to arsenates and oxides (Ehrlich and Newman 2009). WHO (2001) provided a list of these secondary minerals, which Mandal and Suzuki (2002) also tabulated. Moreover, arsenosulfides of almost any metal cation can be found (NRC 1977). Arsenic, which is both chalcophile and siderophile, is found associated with pyrite ( $\text{FeS}_2$ ) or iron hydroxides ( $\text{FeOOH}$ ) and oxides ( $\text{Fe}_2\text{O}_3$  and  $\text{Fe}_3\text{O}_4$ ) in crustal rocks (Nordstrom 2002). Although the maximum As content of pyrite is  $\sim 5\%$ , its typical range is 0.02–0.5% (NRC 1977). Under surface conditions, readily oxidizing As-bearing sulfides and sulfosalts transform to As trioxide and arsenate stages, and subsequently, secondary minerals tend to be arsenates and oxides (NRC 1977). Although igneous rocks show a relatively uniform distribution of As among the major constituent minerals, their sulfide mineral constituents show slight As enrichment (NRC 1977).

Through weathering, As present in sulfides ores are converted into a highly mobile  $[\text{As}(\text{V})]$ , which can enter the As biogeochemical cycle directly in either solid-state as dust particles or through dissolution in the rain, surface, and groundwater (Maity et al. 2011a). Consequently, soil overlying sulfide ore deposits commonly contain several hundred ppm of As, up to 8000 ppm (NRC 1977). In this way, As may be present in unweathered sulfide minerals or in an inorganic anion state. Inorganic arsenate is usually bound to iron and aluminum but could form bonds with any other cation present, viz. calcium, magnesium, lead, zinc (NRC 1977). 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 primarily control groundwater chemistry (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 and siderophile As in contaminated aquifers may result from the dissolution of primary or secondary sulfidic minerals (viz. pyrite), or



desorption from secondary concentrations in hydrous metal oxides (Nordstrom 2002), which is common in the aquifer matrix in many large unconsolidated sedimentary provinces (Mukherjee et al. 2014, 2019).

A simple analysis of the basic types of sulfide ore deposits based on associated geotectonic regimes reveals a systematic pattern. For example, calc-alkaline magmatism at convergent plate boundaries gives rise to (i) Kuroko-type (massive sulfide) deposits in submarine volcanic environments and (ii) Cordilleran-type (postmagmatic) ore deposits in epizonal plutonic settings (Sawkins 1990). Porphyry copper deposits, an important subtype of Cordilleran-type deposits, show a spatial relationship to present or ancient convergent plate boundary regimes (Sawkins 1990). Copper-nickel deposits occurring in ultramafic extrusive or penecontemporaneous intrusive rocks are also related to plate convergence (Sawkins 1990). Sulfide ore deposits generated at divergent plate boundaries (spreading-center systems) are rare as a function of their submarine habitat and are unknown at transform plate boundaries (Sawkins 1990).

On the other hand, stratiform copper deposits and most of the Mississippi Valley-type lead–zinc deposits are formed in continental intraplate environments. In contrast, magmatic deposits in layered mafic complexes also form in intraplate environments, but with spreading-center activity limited in time and space (Masuda 2018). Moreover, gold deposits in greenstone-type volcanic arc sequences can be attributed to a two-stage concentration process at inferred (ancient) convergent plate boundaries (Masuda 2018). Arsenic, being an integral part of all these deposits, implies that primary As distribution is tectonically controlled, transported by secondary processes in solution through leaching or solid-state through physical erosion to the sedimentary basins (see Sect. 5.2).

It is important to emphasize that As contamination, even in a mining district, can sometimes be associated with other lithologies instead of the ore deposits themselves. A study from Osaka Prefecture in western Honshu (Japan) pointed out that although many sulfide mines are present in the Hokusetsu Mountains, they are not essentially sources of As contamination as much as the arsenide disseminated in hornfels and shales (Masuda 2018). On the eastern edge of the Cretaceous–Neogene magmatic bodies in the area, Even et al. (2017) reported high As content in hornfels near the granitic complex that effectively led to As contamination of surface and groundwater. Similarly, As leaching and oxidation in disseminated Miocene sediment-hosted hydrothermal gold (Au) ore deposits led to As contamination of shallow groundwater in Sulawesi, Indonesia (Iskandar et al. 2012). In both cases, the As contamination mechanism was oxidation and decomposition reactions of oxic water infiltrating through a fault system with diffusively distributed sulfides and arsenides (Masuda 2018). Section 6

deals with the basement structure's role in controlling the hydrochemical environment, thus As speciation.

In the mining area of Wassa West district in Ghana, Bhattacharya et al. (2012) found As (besides manganese and iron) contamination in some wells potentially affected by acid mine drainage (AMD) likely to be controlled by the adsorption processes, evident by supersaturation of groundwater with respect to several mineral phases including iron-hydroxides/oxides in the hydrogeochemical models.

## 5.2 Arc Magmas

Reported concentrations of As in various lithologies worldwide suggest a close association with magmatic arcs at convergent continental margins (Fig. 2) and collision belts that inevitably contain arc-derived components (Mukherjee et al. 2014, 2019). Accordingly, high As content in arc magmas can be derived from (a) the subducting oceanic crust with its sediment cover, (b) the mantle component of the subducting slab, (c) the mantle wedge component overlying the slab, (d) the overlying continental crust through which the arc magma ascends to the surface (Hattori and Guillot 2003; Hattori et al. 2005; Guillot and Charlet 2007). Ocean-floor basalts, comprising the oceanic crust are formed by partial melting of the depleted mantle at the mid-oceanic ridge, are not enriched in As (<0.01–1 mg/kg, Deschamp et al. 2013). This implies that As in the oceanic crust is derived from the sediments derived from the continents (terrigenous detritus).

The As concentration in the continental crust is at least two orders of magnitude higher than in the depleted mantle, the source region for mid-oceanic ridge basalts (Matschullat 2000). It is worth mentioning that the arc magma of some volcanoes in the southern part of the Japan-Kurile volcanic chain has low As content, comparable to normal depleted mantle-derived magmas. In contrast, several volcanoes in the northern part of the same region, e.g., Esan (Japan) and Onokotan (Kuriles), which are underlain by thick arc basement, have very high As concentration in their magmas (Noll et al. 1996; Henke 2009). Similarly, medium to high As concentration have been reported from the arc extrusives at continent to ocean convergence-related volcanics of Cordilleran orogenic belts of British Columbia (Boyle et al. 1998), Alaska (Crock et al. 1999), Mexico (Armienta et al. 2001), Chile (Noll et al. 1996) and Perú (Noble et al. 1998). This tells that the As content is almost always high in the settings where the arc magma ascends through an overlying continental crust, implying that interactions of volatile-enriched arc magma with the continental crust is the most important source of primary As enrichment compared to other tectonic settings. This further suggests that (i) in any tectonic regime, the continental crust forms the ultimate

source of groundwater As, (ii) As in continental crust is mobilized hydrothermally by young magmatic hydrous fluids of metasomatic origin, and (iii) As mobilization is enhanced through prolonged interaction between continental crust and hot hydrous fluids or magmas.

Arsenic mobilization from the sediment cover of the subducting plate to the fluid is most effective in continental domains with elevated geothermal gradients, such as near active, e.g., Andes (e.g., Raychowdhury et al. 2014) or ancient magmatic activity centers, e.g., Indus-Tsangpo Suture Zone lithologies of the Himalayas (e.g., Hattori et al. 2005; with up to As 257 mg/kg, Lone et al. 2020). Meteoric water in such domains percolate to depth, becomes heated, and forms hydrothermal systems, e.g., geothermal wells in Costa Rica (As  $\sim$ 25,900  $\mu$ g/L, Hammarlund et al. 2009) that can efficiently remove As and other mobile elements from the crust. The limits of such As mobilization by thermal fluids are controlled and/or governed by the extent of water–rock interaction, which in turn is constrained by the maximum penetration depth of water can (Gleeson and Ingebritsen 2016). Arsenic can be mobilized from deeper parts of the continental crust only if it comes in contact with hot hydrous fluids and gets dissolved. This is evident at active continental margins (mostly hot orogens, Mukherjee et al. 2014, 2019), where hot hydrous fluids mobilize As derived from the subducting slab (Tatsumi 1989). These fluids may or may not be As enriched at the time of slab dehydration. The As content of these fluids depends on the proportions of continent-derived sediments (terrigenous detritus) and hydrated oceanic crust associated with the subducted slab (Mukherjee et al. 2014, 2019). The fluids, which may also be associated with slab-derived melts, subsequently metasomatize the depleted continental lithospheric wedge, lower the mantle solidus and initiate the production of hydrous arc magmas (Mukherjee et al. 2014, 2019). Fractionation of such magmas leads to the generation of high-Al basalts/gabbros, andesites/diorites and dacites/granodiorites (Sisson and Grove 1993) in active or ancient magmatic arcs, which are known to have high to very high (up to 1500 mg/kg) solid-phase As (Henke 2009). The chemistry of the arc magmas may thus be influenced by sedimentary input from the continent into the trench (Plank and Langmuir 1993). At active continental margins, the ascending magma invariably interacts with the (thick) continental crust as it rises, unlike at an ocean–ocean setting; thus, continental arc volcanics tend to have the highest primary As enrichment (Mukherjee et al. 2014, 2019).

As in the Andes, Arc volcanism leads to the deposition of As-rich volcanic ash and other continent-derived sediments (Fig. 2) that can be carried to downstream basins. Arsenic enriched rhyolitic glass, volcanic ash, and associated silicates in sediments and metamorphic deposits, with As up to 10 mg/kg (Raychowdhury et al. 2014), undergo hydrolytic

dissolution and produce an influx of major (e.g.,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Si}^4+$ , and  $\text{HCO}_3^-$ ) and minor (e.g., oxyanions of As, V, and Mo) solutes to groundwater, which leads to precipitation of clays (Mukherjee et al. 2007, 2008; Mukherjee and Fryar 2008; Raychowdhury et al. 2014), under favorable climatic conditions. Himalayan arc-derived lithologies also comprise significant proportions of mafic and intermediate rocks (predominantly andesites), enriched in As compared to their non-arc equivalents, owing to interaction with subduction-zone fluids (Hattori et al. 2005; Guillot and Charlet 2007).

Chemical weathering and leaching of more soluble and labile, solid-phase As from the metastable, high-temperature arc-derived mafic minerals (e.g., olivine, pyroxenes, serpentine) in these rocks (As  $\sim$ 19 mg/kg, Verma et al. 2016) facilitates the release of this As (up to 4100  $\mu$ g/L, Mukherjee et al. 2009a, b) into the hydrological system. Some As-carrying (up to 25,900  $\mu$ g/L (Hammarlund et al. 2009) hydrothermal fluids may also discharge on the surface as hot springs and fumaroles (For more details, see Sect. 5.4 Geothermal and volcanic fluids). Subsequently, with orogeny initiation at active continental margins, the As-bearing arc volcanics coalesce with sediments and rocks of the exhuming orogen. With continuing convergence, slices of serpentinized and, therefore, As-enriched mantle rock (As  $\sim$ 275 mg/kg, Hattori et al. 2005) are also incorporated into the orogen through thrusting (Hattori and Guillot 2003). Sediments derived from the rapidly exhuming orogenic belt by weathering of the As-bearing arc volcanics and obducted, serpentinized mantle rocks are subsequently deposited in adjacent foreland and even back-arc basins (Verma et al. 2016). During protracted convergence over geologic time, multiple generations of foreland basins can form and coalesce with the main orogenic belt, e.g., Siwaliks in the Himalayas (As  $\sim$ 10 mg/kg, Guillot et al. 2015).

Rivers or glaciers cutting through the orogenic belt erode the foreland sediments and exhumed arc derived magmatic rocks (Guillot and Charlet 2007; Verma et al. 2016), and deposit weathered As-rich sediments in the most recent foreland basins, e.g., Terai, Nepal (As up to 27 mg/kg, Guillot et al. 2015). Proximity to these orogenic As sources enhances the possibility of increased chemical weathering and leaching of solid-phase primary As, from meta-stable, high-temperature arc-derived rocks, which constitute primary As source (Verma et al. 2016). With increasing distance from these As sources, weathering and transport lead primary As to remain chemically sorbed mainly in the residual state, more recalcitrant or more strongly bonded, more crystalline or mineralogically mature phases, viz. low-temperature/late-phase silicates (Onishi and Sandell 1955), which are less prone to chemical weathering (Henke 2009). Conversely, with distance, the released As gets physisorbed in more labile secondary solid phases, which act

as the immediate (and secondary) source of groundwater As enrichment in adjoining foreland basin aquifers by redox-dominated reactions (Mukherjee et al. 2014, 2019). This liberated As can co-precipitate with/or get adsorbed onto secondary Fe(III) (oxyhydr)oxides (Smedley and Kiniburgh 2002) or can be re-sequestered by re-sorption or co-precipitation with authigenic Fe(II) sulfides under sulfate-reducing conditions, i.e., aquifer sediment sink (Mukherjee and Fryar 2008; Mukherjee et al. 2008).

To integrate the knowledge of orogen-sourced sediments and groundwater chemistry of ancient (e.g., Himalayan system) and modern (e.g., Andean system) orogens, Mukherjee et al. (2014, 2019) correlated the tectonic settings and groundwater As-enrichment data from 63 major aquifers across the world, to show that a distinctly high proportion of As-affected aquifers are located in convergent (~90%), particularly magmatic arc settings. In stark contrast, only 5% of the basins in other tectonic settings contain groundwater As. However, rift-related basin aquifers are an exception to this generalization (Mukherjee et al. 2014, 2019), although all such rift basins receive magmatic arc detritus, commonly andesite (Peters 2008). Similarly, As-contaminated aquifers in the Red River and the Mekong basins have a mixed convergent/non-convergent setting, with sediment sources from ancient Qamdo-Simao arc rocks (Stanger 2005). Dissolution of volcanic glasses in ash layers and leaching of loess-type deposits in the Chaco-Pampean plain of Argentina have led to groundwater As concentrations from <10 to 5300 µg/L (Nicolli et al. 2012). These examples corroborate that magmatic arcs are the predominant source of primary As in groundwater across the globe.

### 5.3 Allochthonous As-bearing Sediments

The third primary source, preferably a secondary source (see the previous section), of contamination comprises the Cenozoic (mostly Holocene) aquifer sediments. It is worth highlighting that the total As concentration in contaminated groundwater is not as high when compared to the average concentration in soils and sediments, for example, despite total As concentration in Holocene bulk aquifer sediments in Bangladesh ranges from 5 to 16 mg/kg, groundwater has only little over 1 mg/L As (Seddique et al. 2011). This is because groundwater As contamination depends mostly on the chemical form of the As bearing solid phase (mineral species) and changes in the aquatic environment leading to the release of As from aquifer sediments to water (Seddique et al. 2011). The mechanisms for As release from Cenozoic, especially Holocene, aquifer sediments have been described in this section.

As mentioned in the previous section, As-contaminated aquifers are often hosted within extensive clastic sedimentary basins associated with young mountains (Ravenscroft et al. 2009) within or around foreland basins at both active and ancient convergent continental margins (Mukherjee et al. 2014, 2019, 2021), e.g., Indus–Ganges–Brahmaputra basin, the Mekong and Red River deltas in southeast Asia (Alpine–Himalayan Orogenic System), the North American foreland of the western USA (American and Pacific Cordillera) and Canada (Laramide–Nevadan–Sonoma Orogenic System), the Appalachian foreland in New England and adjacent areas of the eastern USA (Appalachian–Ouachita Orogenic System), parts of Argentina, Bolivia, Chile and Peru in the Andean forelands of South America (Andean–Pampean–Toco Orogenic System). Arsenic found in such aquifers can be attributed to the solid phase concentration of As-bearing clasts in their provenance located in major magmatic arcs and orogenic belts at convergent boundaries (Mukherjee et al. 2014, 2019). Thus, As sourced from magmatic arcs may be referred to as primary for both Andean (Ramos Ramos et al. 2012; Raychowdhury et al. 2014; Ormachea Muñoz et al. 2015) and Himalayan (Hattori and Guillot 2003; Hattori et al. 2005; Guillot and Charlet 2007; Mukherjee et al. 2014, 2019; Verma et al. 2016) type orogenic systems, at continental convergent margins across the globe, which after exogenic processes of erosion and transport, ends up in the sediments as a (secondary) source of As, in the aquifers hosted in adjoining foreland basins. Tectonic setting interpretations based on major, trace, and rare earth elements, in conjunction with field measurements of primary sedimentary structures, can help in discriminating provenance and tectonic setting (including ancient, e.g., Proterozoic sedimentary basin in Rajasthan, India, Banerjee and Bhattacharya 1994) of the As-bearing sediments.

### 5.4 Geothermal and Volcanic Fluids

Arsenic contamination of the aquifers—clastic or fractured—might not involve leaching of As from allochthonous clastics or any other host formation lithology. Instead, it might be from As rich geothermal and volcanic fluids that interact with groundwater (e.g., Latin America, Morales-Simfors et al. 2020; Western Iran, Keshavarzi et al. 2011), which are the sources as well as pathways of As. In a recent study covering 15 countries in Latin America (Morales-Simfors et al. 2020), 423 sites were characterized with As originating from geothermal sources, mostly related to present volcanic activity ( $0.001 < \text{As} < 73$  mg/L, mean: 36.5 mg/L) and the transboundary Guarani Aquifer System ( $0.001 < \text{As} < 0.114$  mg/L, mean: 0.06 mg/L).

High As concentrations in geothermal fluids indicate that As is a volatile magmatic component, which is highly soluble in water., e.g., hot springs in Yellowstone National Park, USA have up to 10 mg/L As (Stauffer and Thompson 1984; Webster and Nordstrom 2003), which is released to groundwater and surface water from geothermal fluids within and near the park (Ball et al. 1998, 2002; Nimick et al. 1998). Geothermal fluids in Dominica, the Lesser Antilles, have high As concentrations (>50 µg/L, McCarthy et al. 2005). Moreover, As from hot springs can be mobilized to surface waters and also enter riverbed sediments, e.g., arsenite released from hot springs along the Owens River (USA) gets oxidized to arsenate by microbial activity, and subsequently precipitates and settles to the bottom, thus becoming part of the riverbed sediments with high As content (up to 60 mg/kg, Nimick et al. 1998). It has been observed that geothermal well fluids occasionally contain higher As concentrations (up to 50 mg/L) than associated surface discharges through host springs (Webster and Nordstrom 2003 and references therein). Arsenic content in the outflow (drainage water) from Waikato Power Station, New Zealand was 145 t/year till 2000; however, it has been lowered to ~70 t/year (Webster-Brown et al. 2000 and references therein). This discharge has led to elevated As concentrations (61–1790 mg/kg) in the Waikato River sediments (Webster-Brown et al. 2000).

Geothermal areas occur along plate boundaries, e.g., the Pacific Ring of Fire, in tectonic rift areas, e.g., East Africa, Central India and at seafloor spreading centers, e.g., Iceland, and at hot spots, where mantle-derived plumes ascend, e.g., Hawaii and Yellowstone National Park, USA. Arsenic is one of the incompatible elements, together with antimony (Sb), boron (B), fluoride (F), lithium (Li), mercury (Hg), selenium (Se), and thallium (Tl), and hydrogen sulfide, in high-temperature geothermal settings (Webster and Nordstrom 2003). Moreover, As concentrations are high mainly in thermal waters that leach continental rocks, whereas thermal waters in basaltic rocks, e.g., in Iceland, contain lesser amounts of As. Stauffer and Thompson (1984) demonstrated that As in thermal fluids at Yellowstone National Park in Wyoming, USA, it is derived mainly from leaching host rocks rather than magmas. Arsenic in thermal water is present as [As(III)] in arsenious acid ( $H_3AsO_3$ ). In contrast, in low-sulfide fluids, the [As(III)] in the arsenious acid is oxidized to [As(V)]. The ascending fluid interacts with cold oxygenated groundwater or comes in contact with the atmosphere. On the other hand, in high sulfide solutions, As could be present as thioarsenate complexes (Webster and Nordstrom 2003; Planer-Friedrich et al. 2007).

Since the 1950s, As contamination has been attributed to hydrothermal activity in many geothermal areas of Japan, as reflected by some old works, e.g., Beppu, Kyushu (Kawakami et al. 1956), Tamagawa, Akita, Northeast Japan

(Minami et al. 1958), Hakone, Yugawara, Kanagawa (Awaya et al. 2002), and Toyohira, Hokkaido (Jin et al. 2012). In Kusatsu-Shirane (Gunma), thermal water discharges with up to 7 mg/L of As concentration was reported flowing into the Yukawa River for a decade until the 1990s (Kikawada et al. 2006, 2009). Up to 3.4 mg/L As was reported in thermal water at Kakkonda Geothermal Plant, Akita, Japan (Okada et al. 2002). Acidic (pH 1.2) thermal water with about 2600 µg/L As was reported by Noguchi and Nakagawa (1969) to precipitate out as As sulfides and lead (Pb) As sulfides in surface-water sediments containing ~5–56 wt.% of As. Up to 4.8 mg/L As in thermal waters from geothermal fields in New Zealand are attributed to the leaching of variable composition host rocks. This was considered the mechanism for As enrichment of thermal waters from various geothermal areas in New Zealand.

In Latin America, volcanic and geothermal activity along the convergent plate boundary discharges As-rich waters and gases on the surface in the form of hot springs and fumaroles (Raychowdhury et al. 2014; Mukherjee et al. 2014, 2019, 2021; Coomar et al. 2019). The As concentration of ~50,000 µ/L has been reported by López et al. (2012) in Na-Cl type thermal waters at El Tatio geothermal field in Chile (Landrum et al. 2009). Up to 73,600 µg/L of As concentration has been reported in thermal waters ascending through a wide range of lithologies—sandstones and shales, lava flows and pyroclastics, and metamorphosed carbonate rocks, basalts, and hornblende andesites—in Mexican geothermal fields (López et al. 2012). In the coastal volcanic zones of Central and South America, where the predominant lithological composition is andesitic or rhyolitic, As concentration in thermal waters varies widely, and although As concentrations of several thousand µg/L are frequently reported, none of them is as high as the highest reported As concentration in Mexican geothermal fields (i.e., 73.6 mg/L, López et al. 2012). Reported As concentrations, apparently is contributed by oxidation of sulfide deposits, are as high as 4600 mg/L (?) in the Bolivian Altiplano, where hot springs and fumaroles discharge water and gases in Poopó Lake (López et al. 2012).

In Europe, As concentrations >50 µg/L in groundwater are reported from the Massif Central's geothermal fields in France and in Greece (Brunet et al. 2004; Karydakis et al. 2005). Arsenic concentration in groundwater in Iceland often exceeds 10 µg/L (Arnorrsson 2003), although basaltic lava typically contains less As than silicic lavas (Onishi and Sandell 1955; Baur and Onishi 1969; Ure and Berrow 1982). Geothermal systems in northern and northeastern Spain are characterized by high As concentrations and As-rich mineral deposits around surface geothermal manifestations (Navarro et al. 2011), e.g., hot springs with 50–80 µg/L As in the Caldes de Malavella geothermal field in northern Spain contributes a substantial amount of As (from <1 to



200 µg/L) to groundwater (Piqué et al. 2010). The As concentrations in groundwater (including brines) range from 1.6 to 6900 µg/L in the Phlegraean geothermal fields in southern Italy (Aiuppa et al. 2006). Quaternary volcanic rocks with hydrothermal activity on Ischia (a volcanic island offshore from Naples, Italy, in the Tyrrhenian Sea) contribute As to groundwater at concentrations ranging up to 3800 µg/L (Aiuppa, et al. 2006; Daniele 2004).

In the Middle East, reported As concentrations in thermal waters are as high as 3500 µg/L near Mt. Sabalan, a stratovolcano in northwestern Iran (Haeri et al. 2011). In western Anatolia, Turkey, natural leaching, aided by pumping and discharge of waste geothermal fluids from an active geothermal system, has mobilized As from metamorphic, igneous, and sedimentary rocks to groundwater in a shallow alluvial aquifer, where highest As concentrations in groundwater and geothermal fluids were 561 and 594 µg/L, respectively (Gunduz et al. 2010). Baba et al. (2021) reported that geothermal fluids circulating through volcano-sedimentary units in the Anatolian plate caused a more effective alteration, increasing As concentrations in geothermal systems. Moreover, they determined that high As concentrations within the convective-non-magmatic extensional geothermal play types, viz. Western Anatolian Extensional System and the North Anatolian Fault. Bundschuh et al. (2013) found the mixing of geothermal fluids primarily responsible for contamination of freshwater resources, making them unsuitable for drinking or irrigation, based on hydrogeochemical investigations of geothermal fluids from deep wells and hot springs in western Anatolia, Turkey. Baba and Sözbilir (2012) deciphered that the enhanced dilation ~ E–W-strikes of the graben faults in west Turkey, accommodating the deep circulation of hydrothermal fluids of meteoric origin, provide a suitable environment for the presence of high As levels in geothermal fluids.

Groundwater in early Proterozoic silicic volcanics and granites of the Chhattisgarh Basin (India) contains >10 µg/L of As, emplaced there by hydrothermal fluids (Acharyya 2005).

Geothermal systems are controlled by global tectonic and local structural settings, which ultimately define heat and fluid flow regimes. See the chapter Tectonic and Structural Control on Geothermal Systems in the same book for more details. This association of geothermal systems with geotectonic settings advocates for geotectonic control in distributing and disseminating As.

## 5.5 Coal and Hydrocarbon Resources

Together with both of these fuel resources' exploitation processes, coal deposits and hydrocarbon reservoirs are

other geogenic sources of As contamination that is accelerated by the extraction and refining processes and subsequent usage. Geotectonics also controls their formation in ancient and modern sedimentary basins. The average world concentration of As in coal is  $9.0 \pm 0.8$  and  $7.4 \pm 1.4$  mg/kg for bituminous coals and lignites, while in their ash, it is  $50 \pm 5$  and  $49 \pm 8$  mg/kg, respectively (Yudovich and Ketris 2005).

Petroleum reservoir waters from sedimentary basins can contain As as a result of the evaporation of seawater that concentrates As in oil reservoirs (Birkle et al. 2010). For example, Birkle et al. (2010) reported As concentration up to 2000 µg/L at depths of 2900–6100 m below sea level in oil fields of southeastern Mexico. Moreover, formation water (fresh or saline water trapped in geological formations for thousands to millions of years) forms the most significant part of the volume of co-produced water in hydrocarbon production (Neff et al. 2011). Stromgren et al. (1995) found that the formation water has particularities of the geological formation and is polluted by metals, inorganic compounds, and minerals, including As. Also, their concentrations can differ considerably from seawater.

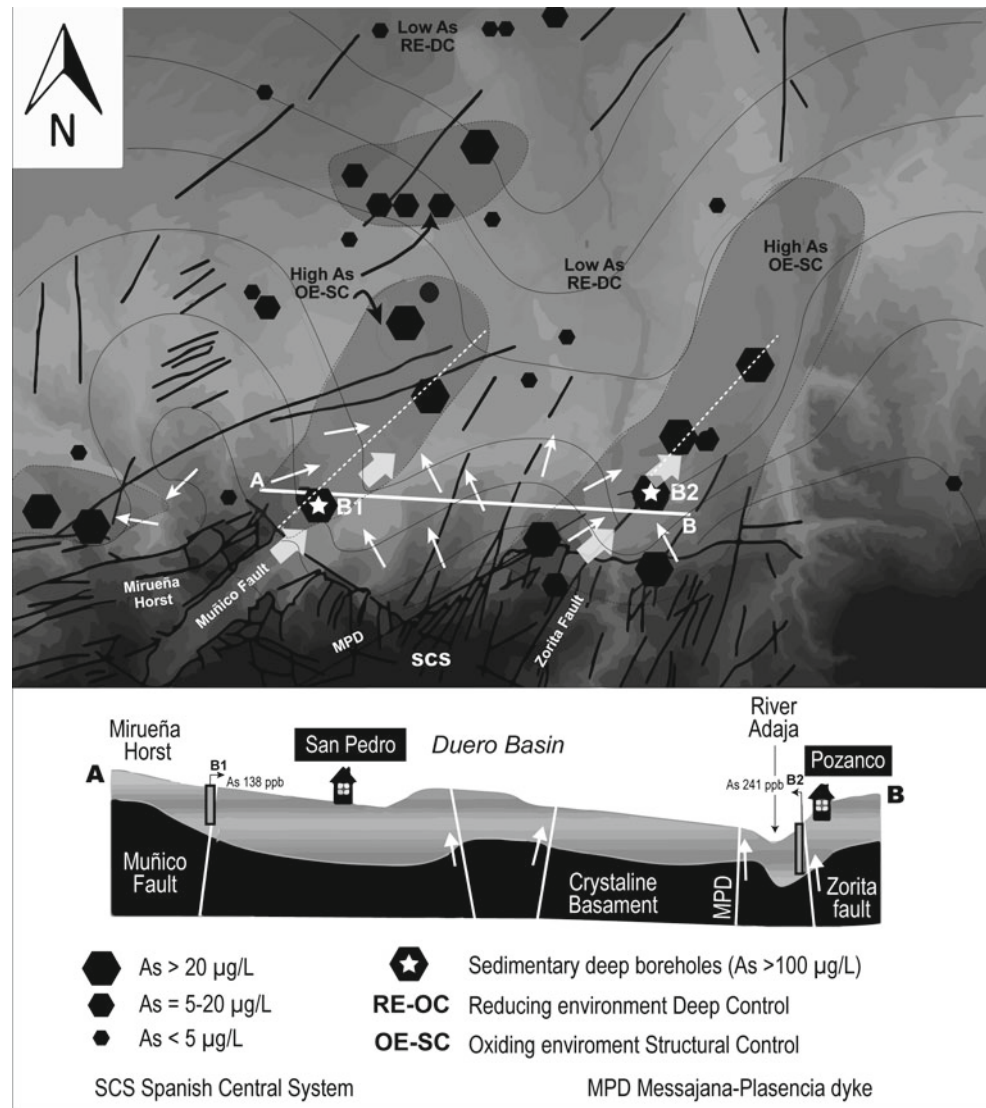
## 6 Structural Controls on As Concentration and Distribution in Aquifers

Apart from the aforementioned solid and liquid phase inputs of As, attributed to the lithology and geothermal and volcanic fluids, respectively, there is yet another essential aspect of As enrichment of aquifers, which has to do with the structural settings of the aquifers in a basin, e.g., Duero Basin, Spain (Giménez-Forcada and Smedley 2014), wherein basin morphology, prolongation of faults, especially division of the underlying basement into blocks, produces discrete hydrodynamic environments, resulting in spatial variations in hydrochemical composition, including As speciation (Fig. 4, Giménez-Forcada and Smedley 2014).

Although pH and redox conditions play essential roles in controlling the As occurrence, distribution, and mobility in groundwater, the basement structure plays a significant control, e.g., faults behave as effective conduits for groundwater flow and influence the recharge of the basin (Giménez-Forcada and Smedley 2014). The groundwater flow could be controlled by the faults (or fault zones), which act as preferential flow conduits and zones in tectonic depressions (Vilanova et al. 2008), particularly those faults oriented parallel to the general flow path in the basin (Giménez-Forcada and Smedley 2014). Moreover, horst and graben structures can generate discrete hydrogeological cells (Giménez-Forcada and Smedley 2014).

Groundwater chemistry varies laterally and with depth, depending upon climatic and hydrological factors, viz.

**Fig. 4** Groundwater (springs and boreholes) Arsenic distribution, along with the prominent structural fractures mapped by Simón Gómez (1996), in Duero Basin. Deep boreholes B1 and B2 with the highest As concentrations are represented in the cross-section A–B. Blue contours indicate the piezometric surface. Symbol size reflects arsenic concentration. Modified after Giménez-Forcada and Smedley (2014)



evaporation (Giménez-Forcada and Smedley 2014). For example, in the shallow sedimentary aquifer (<40 m), evaporation can increase the concentrations of dissolved elements, including trace elements like As (e.g., Fujii and Swain 1995; Welch and Lico 1998; Smedley and Kiniburgh 2002). For shallow wells (<40 m) in Duero Basin, Spain, oxidizing conditions and evaporation control groundwater composition. At 40–160 m depth, reducing conditions affect the concentrations of As and other redox-sensitive species, viz.  $\text{SO}_4$ ,  $\text{NO}_3$ , Fe, and Mn (Giménez-Forcada and Smedley 2014). The low As concentration at depth is consistent with As substitution into sulfide minerals under reducing conditions (Giménez-Forcada and Smedley 2014).

However, in deeper boreholes within the sedimentary aquifer (200–450 m) in Duero Basin, the dominant waters are  $\text{Na-HCO}_3$  type with alkaline pH (>9) and high As (up to

241 µg/L, borehole B2), which implies the presence of an oxidizing environment (Fig. 4, Giménez-Forcada and Smedley 2014). These authors explained it in terms of the contribution of oxidizing flows following the main direction of faults in Duero Basin. These deep flow paths (coming directly from the basement and also through the discontinuities within the sedimentary basin) can also transport dissolved As from other As-enriched zones under oxic and alkaline groundwater conditions (Giménez-Forcada and Smedley 2014).

The basement structures consisting of an alternation of horsts and grabens, as in the case of Duero Basin, which is associated with late or post-Variscan tectonism (Ubanell 1985; Ares Yañez et al. 1995; Gómez Ortiz and Babín Vich 1996; Simón Gómez 1996; Herrero 1999; Giménez-Forcada and Smedley 2014), have their manifestation on the surface as large lineaments or areas with a high density of smaller

lineaments (Fig. 4) and define subsequent sedimentation and lithological discontinuities (Colmenero et al. 2001; Armenteros et al. 2002; Alonso Gavilán et al. 2004), which in turn control the hydrogeological and hydrochemical features in the basin (Alberruche Del Campo 2010).

The major fault structures thus control the occurrence of groundwater As, as in Duero Basin (Giménez-Forcada and Smedley 2014), where groundwater extracted from the deep boreholes along the main fault lines, taking advantage of the direction of preferential recharge flows, are oxidizing, alkaline, Na-HCO<sub>3</sub> type with relatively high concentrations of As, up to 241 µg/L As, albeit with limited data, appear to have high percentages of [As(V)], along with U, V, and F (Giménez-Forcada and Smedley 2014).

Another structural control could be the hydraulic gradient, the lower one providing more residence time for groundwater (Bhattacharya et al. 2002; Nriagu et al. 2007). Smedley and Kinniburgh (2002) pointed out that whether released As remains at problematic levels in groundwater depends not only on whether there are biogeochemical reactions that retard the transport of As but also upon the hydrologic and hydrogeologic properties of the aquifer, such as flow velocity and dispersion. If the kinetics of As release is slow, and groundwater residence time is short, then As concentrations may not increase to the point where groundwater would be considered contaminated. Conversely, with rapid As mobilizing reactions and prolonged residence, As can accumulate in groundwater such that concentrations become hazardous, e.g., Bangladesh (Zheng et al. 2004; Bhattacharya et al. 2009). Eventually, if the biogeochemical conditions that lead to As release and mobilization continue to be present (within a geologic timeframe), the source could become exhausted.

Bhattacharya et al. (2012) highlighted the importance of groundwater divides, which leads to many local flows, thus forming different groundwater systems controlling As and other toxic element concentrations in Wassa West district's mining area in Ghana. The aquifers in this hilly area are not affected severely by the leaching of minerals because of short groundwater residence time and intense flushing because of the high hydraulic gradient. Thus, groundwater flow systems' local character prevents AMD's significant impact on groundwater systems on a regional scale.

## 7 Conclusions

Based on the average crustal concentration and presence in different geological materials, any aquifer is potentially susceptible to contamination. Instead, there is heterogeneity in the level of As contamination of groundwater across the globe, albeit with a pattern defined mainly by geotectonics. For example, major occurrences of contaminated aquifers

within sedimentary basins located in the vicinity of active or ancient magmatic arcs at continental convergent margins are controlled by geodynamic processes operating at a regional-scale over millions of years. They are not mere manifestations of heterogeneous As-mobilizing (bio) geochemical conditions.

As enrichment of aquifer matrix in the forelands basins and subsequently the residing groundwater therein is also related to exhumation and erosion of arc-related lithologies that supply As-enriched sediments to these basins. Although rift basins seem to be apparent exceptions to the aforementioned conclusion of associating As contamination to convergent boundaries; however, those basins also derive sediments, at least substantially if not alone, from nearby magmatic arcs. So this observation regarding As contamination of aquifers in the rift basin does not contradict the previous conclusion. Arsenic contamination is widespread globally; however, contaminated areas tend to concentrate in and around active tectonic zones (modern/active and ancient), manifested along with previously mentioned arc magmatism, through volcanics center, geothermal fields, mineralized zones (especially metal sulfides), etc.

Among geogenic sources of As contamination, the most common occurrences are metal sulfides and sorbed species on Fe hydroxides, although As occurs within some silicate and carbonate rocks as well. Although geotectonics control the primary geologic sources of As through regional scale geodynamic processes, the basement structure of the basin, also driven by (paleo)geotectonics, may control As speciation locally through the development of compartmentalized hydrodynamic environments, resulting in spatial variations in hydrochemical composition, including As speciation.

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