



Research article



Arsenic in a groundwater environment in Bangladesh: Occurrence and mobilization

Md. Enamul Huq^a, Shah Fahad^{b,c,**}, Zhenfeng Shao^{a,*}, Most Sinthia Sarven^b, Imtiaz Ali Khan^c, Mukhtar Alam^c, Muhammad Saeed^c, Hidayat Ullah^c, Muahmmad Adnan^c, Shah Saud^d, Qimin Cheng^e, Shaukat Ali^{f,g}, Fazli Wahid^c, Muhammad Zamin^c, Mian Ahmad Raza^c, Beena Saeed^c, Muhammad Riaz^h, Wasif Ullah Khanⁱ

^a State Key Laboratory of Information Engineering in Surveying, Mapping and Remote Sensing, Wuhan University, 129 Luoyu Road, Wuhan, 430079, China

^b College of Plant Science and Technology, Huazhong Agricultural University, Shizishan Street-1, Wuhan, 430070, Hubei, China

^c Department of Agriculture, University of Swabi, Khyber Pakhtunkhwa, Pakistan

^d Department of Horticulture, Northeast Agriculture University, Harbin, China

^e Huazhong University of Science and Technology, School of Electronics Information and Communications, 1037 Luoyu Road, Wuhan, 430074, China

^f Global Change Impact Studies Centre (GCISC), Ministry of Climate Change, Pakistan

^g Environmental Monitoring and Science Division, Alberta Environment and Parks, Canada

^h Department of Environmental Sciences and Engineering, Government College University Faisalabad, Allama Iqbal Road, Faisalabad, Pakistan

ⁱ National Engineering Laboratory for Tree Breeding, College of Biological Sciences and Biotechnology, Beijing Forestry University, Beijing, 100083, China

ARTICLE INFO

Keywords:

Arsenic
Geochemical process
Arsenic sources
Arsenic mobilization
Bangladesh

ABSTRACT

Groundwater with an excessive level of Arsenic (As) is a threat to human health. In Bangladesh, out of 64 districts, the groundwater of 50 and 59 districts contains As exceeding the Bangladesh (50 µg/L) and WHO (10 µg/L) standards for potable water. This review focuses on the occurrence, origin, plausible sources, and mobilization mechanisms of As in the groundwater of Bangladesh to better understand its environmental as well as public health consequences. High As concentrations mainly occur from the natural origin of the Himalayan orogenic tract. Consequently, sedimentary processes transport the As-loaded sediments from the orogenic tract to the marginal foreland of Bangladesh, and under the favorable biogeochemical circumstances, As is discharged from the sediment to the groundwater. Rock weathering, regular floods, volcanic movement, deposition of hydrochemical ore, and leaching of geological formations in the Himalayan range cause As occurrence in the groundwater of Bangladesh. Redox and desorption processes along with microbe-related reduction are the key geochemical processes for As enrichment. Under reducing conditions, both reductive dissolution of Fe-oxides and desorption of As are the root causes of As mobilization. A medium alkaline and reductive environment, resulting from biochemical reactions, is the major factor mobilizing As in groundwater. An elevated pH value along with decoupling of As and HCO₃⁻ plays a vital role in mobilizing As. The As mobilization process is related to the reductive solution of metal oxides as well as hydroxides that exists in sporadic sediments in Bangladesh. Other mechanisms, such as pyrite oxidation, redox cycling, and competitive ion exchange processes, are also postulated as probable mechanisms of As mobilization. The reductive dissolution of MnOOH adds dissolved As and redox-sensitive components such as SO₄²⁻ and oxidized pyrite, which act as the major mechanisms to mobilize As. The reductive suspension of Mn(IV)-oxyhydroxides has also accelerated the As mobilization process in the groundwater of Bangladesh. Infiltration from the irrigation return flow and surface-wash water are also potential factors to remobilize As. Over-exploitation of groundwater and the competitive ion exchange process are also responsible for releasing As into the aquifers of Bangladesh.

* Corresponding author.

** Corresponding author. College of Plant Science and Technology, Huazhong Agricultural University, Shizishan Street-1, Wuhan, 430070, Hubei, China.

E-mail addresses: shah_fahad80@yahoo.com (S. Fahad), shaozhenfeng@whu.edu.cn (Z. Shao).

<https://doi.org/10.1016/j.jenvman.2020.110318>

Received 20 January 2019; Received in revised form 16 June 2019; Accepted 20 February 2020

Available online 3 March 2020

0301-4797/© 2020 Elsevier Ltd. All rights reserved.

1. Introduction

Arsenic (As) is a ubiquitous element and is ranked 20th in abundance within the earth's crust, ranked 14th in seawater, and ranked 12th in the human body (Flora, 2015). As and its compounds are transportable in surface and subsurface environments. The weathering of rocks converts arsenic sulfide into arsenic trioxide. As is a toxic element that normally originates in subsurface water and is one of the major important groundwater contaminants. Thus, As comes enters As cycle as dust and/or by dissolution in rain, open water (such as rivers, ponds) and groundwater (Chowdhury et al., 2018; Cui et al., 2018; Minatel et al., 2018). Therefore, As contamination in groundwater is a severe risk to humans throughout the world. As can penetrate into the food chain through plants and animals. As a result, living organisms (plant and animal) throughout the world are exposed to the toxicity of As. Initially, As is spread by air, food, and water (both surface and ground) and then enters the human body (Ahmed et al., 2016a; Zhu et al., 2019; Sönmez et al., 2016; Turan et al., 2017, 2018; Zahida et al., 2017). Naturally occurring inorganic As is the major source of As in Bangladesh (Alam and McPhedran, 2019; Bose and De, 2013; Islam et al., 2017b). However, more and long-term inclusion of As with food along with water causes arsenicosis which is responsible for many serious diseases (Bhowmick et al., 2018; Rahman et al., 2018a; UNICEF, 2007). It is projected that approximately 100 million people throughout the world are at risk of As contamination; interestingly most of those at risk are from developing countries. To date, arsenicosis has become the largest chemical threat to the public health (Sharma et al., 2014). Recently, the large amount of As poisoning in the South East Asian nations has become a major socioeconomic and health crisis (Argos et al., 2007; Bhattacharjee et al., 2019; Garshick et al., 2017; Khan et al., 2009; Mahmood and Halder, 2011).

Since the 1970s, As contamination in the environment (human and physical) has been reported worldwide (Singh et al., 2015). As occurrence in drinking water has risen across geographic borders and has caused a global environmental and socio-economic threat. Occurrence of high As in drinking water is considered a serious health hazard all over the world (Agusa et al., 2014; Ahmed et al., 2016b; Barats et al., 2014; Bortnikova et al., 2012; Brima et al., 2006; Dousova et al., 2016; Fang and Chen, 2015; Fransisca et al., 2015; Li et al., 2016; Maizel et al., 2016; Martin et al., 2015; Martínez-Villegas et al., 2013; Pell et al., 2013; Pi et al., 2016; Su et al., 2016; Wang et al., 2009; Xie et al., 2012b). Recently, the environmental fate and behavior of As have become a focus in southeast Asia. Bangladesh and West Bengal, India, are mostly invaded and vulnerable areas due to the elevated As concentration in groundwater (Banerjee et al., 2017; Rahman et al., 2015). According to a World Health Organization (WHO) report, As occurrence in the groundwater of Bangladesh is "the largest mass poisoning of a population in history" (Singh et al., 2015). However, Bangladesh is composed of four principal geomorphologic regions, namely, a Flood plain, Deltaic, Fig. 1 and Hill tract. Water analysis demonstrates that the hand tube-wells from the Fig. 1 and as well as Hill tracts are mostly free of As contamination, but the Flood plains, Deltaic regions, and Coastal areas are highly As-contaminated (Chakraborti et al., 2010). The groundwater of 50 out of 64 districts (Fig. 1) contains As exceeding the Bangladesh standard (50 µg/L). Moreover, 59 districts contained a higher content of As than the WHO recommended value (10 µg/L) for As in potable water (Chakraborti et al., 2015). So far, the government of Bangladesh is not very aware of As pollution problem. For example, the WHO has recommended the limit of groundwater As to be a standard of 10 µg/L worldwide and for Bangladesh, 50 µg/L (Goel et al., 2019; Kipler et al., 2016). However, approximately 57 million people world-wide and 35 million people in Bangladesh are drinking water with an As concentration above the WHO and Bangladesh standards (Bodrud-Doza et al., 2016).

However, several research studies related to As contamination and its risk to human health, society and the environment as well as the

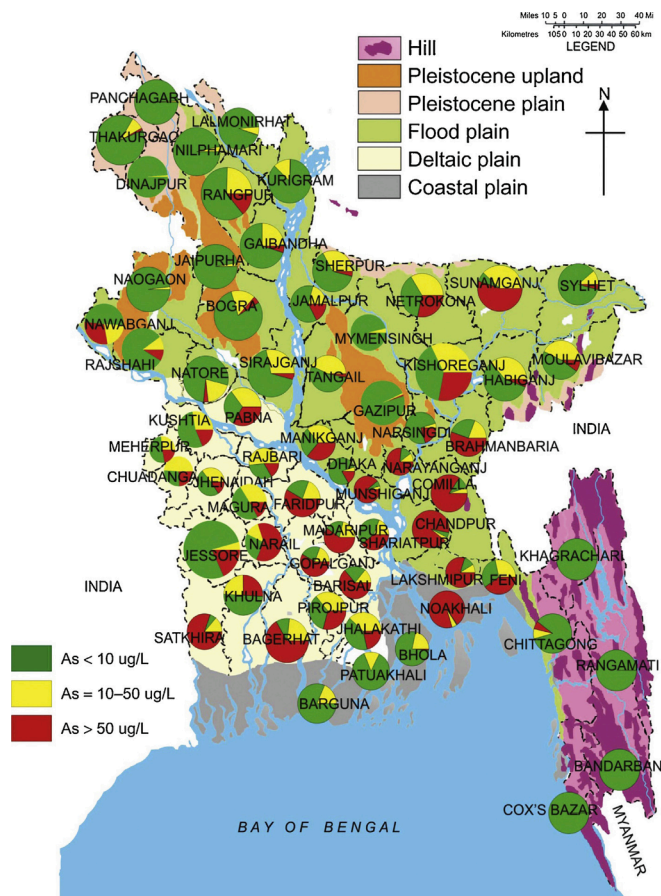


Fig. 1. As distribution of all geo-morphological regions in Bangladesh (Chakraborti et al., 2015).

hydrogeochemistry and geochemical processes of As enrichment and mobilization in the groundwater in Bangladesh have been performed (Ahmad et al., 2007; Ahmed et al., 2004; Ahmed et al., 2016a; Alam and McPhedran, 2019; Argos et al., 2007; Dhar et al., 1997; Dowling et al., 2002; Harvey et al., 2006; Hasan et al., 2009; Huq et al., 2019; Joseph et al., 2015a, b). Nevertheless, a few systematic review studies have been performed regarding the As enrichment and mobilization processes in the groundwater environment in Bangladesh. Moreover, the origins of As in the groundwater environment are well understood for many areas across the world. Therefore, a more detailed study is required to characterize the complex processes of As enrichment and mobilization in groundwater aquifers and its matrix in Bangladesh. The objective of the current review is to provide an extensive study of the different processes as well as the factors responsible for the enrichment of As and for controlling the geochemical routes for As mobilization in the groundwater environment in Bangladesh. This review will help the researchers to discover the specific reasons for As the enrichment and mobilization processes in the groundwater in Bangladesh.

2. Chemical form of Arsenic

In natural environment As is a ubiquitous heavy metal and extremely toxic for the all living organisms. It is a crystalline "metalloid", and a natural constituent, characterized by in-between metals as well as nonmetals (Mandal and Suzuki, 2002). It exists mostly within four oxidation forms, namely arsenic [As(0)], arsenate arsenite [As(III)], arsine [As⁻(III)], and [As(V)]. The solubility of it depends on the pH along with ionic environment. Among the four forms of As, the As(V) is the most durable form (Gupta et al., 2011; Sharma and Sohn, 2009; Zhao et al., 2010). Thermodynamically As(V) is stable in the aerobic water,

whereas As(III) is predominantly reduced in redox environment. As can be found in environment with different chemical forms for instance arsenobetaine $[(\text{CH}_3)_3\text{As} + \text{CH}_2\text{COOH}]$, monomethylarsonic acid $[\text{CH}_3\text{AsO}(\text{OH})_2]$, trimethylarsine oxide $[(\text{CH}_3)_3\text{AsO}]$, dimethylarsinic acid $[(\text{CH}_3)_2\text{As}(\text{OH})]$, arsenolipids, arsenocholine, arsenosugars, etc. As (III) is typically more poisonous than that of As(V) (Abedin et al., 2002). Moreover the dimethylarsinous acid as well as monomethylarsonous acid are much toxic than that of their original compounds (Mass et al., 2001; Petrick et al., 2000). However, there are four majors As species are occurred in water with two inorganic forms such as [arsenite (H_2AsO_3^-) and arsenate (H_2AsO_4^-)]. These types of As are usually available in groundwater of Bangladesh. Another two are found in organic forms for instance monomethylarsonic acid $[\text{CH}_3\text{AsO}(\text{OH})_2]$ as well as dimethylarsinic acid $[(\text{CH}_3)_2\text{As}(\text{OH})]$ (Hossain, 2006; Singh et al., 2015).

3. Evolution and occurrence of Arsenic in Bangladesh

The As contamination in Bangladesh groundwater was first proved by DPHE (Department of Public Health Engineering) in the district of Chapai Nawabganj in late 1993 (BGS and DPHE, 2001). Some 23,000 tube-wells were randomly checked by the staff of DPHE to measure the level of As, and they found that the degree of the problem was very severe and identified that the centre part of the southeast Dhaka, capital of Bangladesh, was the worst affected area (Chakraborti et al., 2015). The northern parts of Bangladesh were found to be an 'uncontaminated' region in consideration of the prevailing As concentration of the Bangladesh standard (50 $\mu\text{g}/\text{L}$) for drinking water. However, a number of hot spots were found in Northern Bangladesh since As patients were identified in this area. Subsequent testing of the tube-wells established the overall emergent pattern of the threat of As in the groundwater in Bangladesh. The shallow aquifer (<50 m) of Bangladesh produces highly As-contaminated groundwater. Several studies have demonstrated that the shallow aquifers from different parts of Bangladesh are badly affected by the high levels of As poison; comparatively, the deep aquifers remain safe from As contamination (<10 $\mu\text{g}/\text{L}$) (Duan et al., 2017; Huq et al., 2018; McArthur et al., 2001).

However, the earliest incidence of As pollution in the groundwater of West Bengal, India, was observed in 1984 (Bhowmick et al., 2018). In 1983, As poisoning in this area was first identified by K. C. Saha from the Department of Dermatology, School of Tropical Medicine. From the early 1980s, approximately 1068 As-related patients in West Bengal were observed because patients regularly used As-contaminated groundwater from tube-wells for drinking as well as other household purposes. Later, the School of Environmental Studies (SOES), Jadavpur University Calcutta, India, initiated their As field survey in 1988 and identified more than 600 As patients with skin lesions (Chakraborti et al., 2002). However, the severity of As poison in the groundwater of Bangladesh might be as intense as the severity of As poison in the groundwater of West Bengal. Therefore, the SOES informed the WHO and UNICEF (United Nations International Children Emergency Fund) as well as the government of Bangladesh in early 1994 regarding the probability of widespread As contamination in the groundwater in Bangladesh (van Geen et al., 2014). The international conference related to As was organized by SOES in Calcutta in February 1995 (Galbacs and Galbacs, 1995), and the delegates of the WHO, representatives of UNICEF in Bangladesh as well as government officials from Bangladesh were in attendance at this conference. However, in that conference, on one reported on the As pollution in the groundwater of Bangladesh (Chatterjee et al., 2010; Islam et al., 2017e). Immediately after that international conference, some medical personnel of Bangladesh were contacted and notified SOES that for several years they had been dealing with skin lesion patients who were similar to the patients of West Bengal in the out-patient departments in their hospitals. In addition, the doctors admitted that during the conference period they were not aware that these types of skin lesions occurred as a result of As toxicity. Since 1995, the number of people with skin lesions has increased in

Bangladesh, predominantly from districts such as Faridpur, Cahnspur, Narayanganj, Sathkhira, and Bagerhat. The patients from these districts started going to Calcutta, India, to receive treatment. Moreover, some of the patients carried hand tube-well water samples for As analysis. Interestingly, SOES found that most of the samples were highly As-concentrated (Hassan et al., 2005; Mahmood and Halder, 2011; Parvez et al., 2017; van Geen et al., 2014).

In 1996, the Department of Geology and Mining, University of Rajshahi, Bangladesh, sent some 600 groundwater samples to SOES collected from 4 districts (i.e., Rajshahi, Kushtia, Nawabganj, and Jessore) to be analyzed for the presence of As. Most of those groundwater samples were detected as having elevated As contamination (Dhar et al., 1997; Rasul et al., 2002; Reza and Jean, 2012). In addition, two medical staff members of the National Institute for Preventive and Social Medicine (NIPSOM) were sent by the WHO to Bangladesh as well to the SOES to receive training for two weeks in June 1996 to understand the characteristic symptoms of arsenicosis. Those medical staff members also brought some hand tube-well water as well as biological samples from the affected districts from Bangladesh and found high levels of As in the samples. Subsequently, SOES and the NIPSOM worked together in Bangladesh and surveyed some districts. Approximately 750 groundwater samples from tube-wells along with approximately 300 hair, nail and skin samples were gathered from As-affected regions for As analysis. The analyzed samples were positive for elevated As contamination (Anwar et al., 2002; BBS and UNICEF, 2011; BGS and DPHE, 2001; Kinniburgh et al., 2003). Although the As contamination in the groundwater of Bangladesh was initially found in 1993, a detailed survey of As contamination in groundwater was not started until 1996 (Dhar et al., 1997).

During 1998, an International Conference related to As was held in Dhaka, Bangladesh. After this conference, the scale of the groundwater As contamination in Bangladesh was exposed (Chakraborti et al., 2002). Throughout this conference, the SOES and Dhaka Community Hospital (DCH) collected approximately 8065 groundwater samples from hand tube-wells covering 60 districts across Bangladesh and noted that 66% of the tube-well water samples showed As exceeding 10 $\mu\text{g}/\text{L}$, which is the WHO guideline value for drinking water, and that 51% had As higher than 50 $\mu\text{g}/\text{L}$ (Bangladesh standard) (Chakraborti et al., 2015). In addition, they also revealed that the As concentration of groundwater samples of 52 districts was higher than 10 $\mu\text{g}/\text{L}$ and that for 41 districts was more than 50 $\mu\text{g}/\text{L}$. Consequently, the British Geological Survey (BGS) and the DPHE during March–June of 1998 and May–July of 1999 conducted another survey. During those surveys, they collected 3534 tube-well samples from all over the country (without Chittagong Hill tracts). After they analyzed the collected samples, they showed that 46% of the tube-wells had As above 10 $\mu\text{g}/\text{L}$ and 27% exceeded 50 $\mu\text{g}/\text{L}$ (BGS and DPHE, 2001). In addition, BGS and DPHE (2001) were informed that approximately 57 million world residents and 35 million residents of Bangladesh may be drinking As-polluted water with higher than the WHO guidance standard and the Bangladesh standard, respectively. Then, several surveys were conducted to analyze the magnitude of As in hand tube-well water over the country using field testing kits with the endeavors of a number of organizations, namely, the Bangladesh As Mitigation and Water Supply Project, Danish International Development Agency, DPHE/UNICEF, World Vision International and the Asia Arsenic Network, which were monitored by the National Arsenic Mitigation Information Centre. According to their database, it is estimated that 1.4 of 4.8 million (approximately 30%) tube-wells of Bangladesh had As higher than 50 $\mu\text{g}/\text{L}$ (Harvey et al., 2006). In Bangladesh, almost 20% of the shallow tube-wells contain As exceeding 50 $\mu\text{g}/\text{L}$, which is more than the Bangladesh standard. Some 8000 villages of the country where 80% of the hand tube-wells are As-contaminated showed 50 $\mu\text{g}/\text{L}$ (Chakraborti et al., 2015).

The As concentration in the groundwater in Bangladesh has been found to be widely range (<0.5->4600 $\mu\text{g}/\text{L}$) (Alam et al., 2003; Bhattacharya et al., 2009; Bibi et al., 2008; Chowdhury et al., 2018; Harvey

et al., 2006; Radloff et al., 2007, 2017; Reza and Jean, 2012; van Geen et al., 2014; Whaley-Martin et al., 2017). In Bangladesh, approximately 25% of the tube-wells are reported to contain As above 50 µg/L; even 42% of the tube-wells exceeded the level of As of 10 µg/L (Mosler et al., 2010). The Southern along with the Eastern regions of the country are the most badly As-contaminated regions, with almost 90% contaminated tube-wells. However, in some districts, the number of As-contaminated tube-wells is very low (approximately 1%), and these districts are located in the Pleistocene upland of Madhupur along with the Barind tracts in Northern Bangladesh. Various As-contaminated 'hot-spots' are observed in some of the Northeastern, Northwestern, Southwestern and North-central portions of the country (Huq et al., 2018b; Islam et al., 2017f; Kanoua and Merkel, 2017; Majumder et al., 2013; Reza et al., 2010a; von Brömssen et al., 2014). More than 60% of the tube-wells of the Ganges-Brahmaputra flood plains and deltaic plains are As-contaminated, whereas 80% of the tube-wells from the shallow aquifer of the Meghna River catchment, including the coastal plain, are heavily As-contaminated. However, the Southern deltaic plains within the Ganges, Brahmaputra and Meghna (GBM) Rivers catchment of Bangladesh are excessively As-contaminated, more than the flood plain of northern parts of Bangladesh (Das et al., 2018; Edmunds et al., 2015; Hasan et al., 2009; Jung et al., 2015; Shamsudduha et al., 2008; Verma et al., 2015; Yu et al., 2015). The As concentration shows an extensive range in spatial inconstancy, even at the local level (tube-well to tube-well), that makes it too hard to forecast As concentration for the unsampled tube-wells, even with the information of the As concentration of the adjacent tested tube-wells (Chowdhury et al., 2018; Fendorf et al., 2010; Hassan et al., 2003; Islam et al., 2017a, 2017c; Pi et al., 2016).

The occurrence of As also varies based on depth. Highly As-contaminated groundwater in Bangladesh is observed at a depth of 38 m, indicating transportation and release of As at shallower depths, as suggested previously (Aziz et al., 2017; Kanoua and Merkel, 2017; Munoz et al., 2013; Ramos et al., 2014). As discharge at this depth might also occur because of the low adsorption ability of the Fe-sulfide minerals (Cao et al., 2018). Most of the shallow aquifers (<50 m) in Bangladesh are producing highly As-contaminated groundwater. The shallow aquifers of Bangladesh are badly affected by high As compared to the deep aquifers. The deep aquifers still remain safe from the As poison (Michael and Khan, 2016; Mosler et al., 2010; Mukherjee and Fryar, 2008). The shallow groundwater (10–50 m) of the delta aquifers is mostly composed of unconsolidated sediments and has a high As concentration (Radloff et al., 2017; Wang et al., 2019). However, the complexity of As occurrence depends on the pattern of both depth as well as geological setting of a certain area (Guo et al., 2017; Hasan et al., 2009; Hussain and Abdullah, 2001; Kinniburgh and Smedley, 2001; von Brömssen et al., 2014). The differences in As concentrations within shallow and deep aquifers could be attributed to the total As concentration in the sediment. In addition, As in the sediment also depends on the size of the grain, the capability of sorption regarding the grains of the sediment, the condition of redox in the aquifers, the duration of the groundwater flushing, as well as the circumstances of artificial withdrawal of groundwater (Freikowski et al., 2013; Lin et al., 2017; McArthur et al., 2001; Schaefer et al., 2017; Stollenwerk et al., 2007).

4. Groundwater Arsenic and the public health threat

As exposure to humans occurs via ingestion, breathing, dermal contact, as well as genetically. Mostly, diet is the major source of As exposure for humans, while ingestion from air and soil is normally a much smaller source (Khan et al., 2009; Ourshalimian et al., 2019). In Bangladesh, the worst effects of As on human health were first observed in 1993 (Kile et al., 2016; Smedley and Kinniburgh, 2002). The common health consequences of As include hindrance in the development of children, skin disorders (e.g., melanosis, hyperkeratosis, leucomelanosis, keratosis, even skin cancer), hypertension, non-pitting edema,

gangrene, intestinal cancers (i.e., bladder, kidney, and lung), tangential vascular disease, ischemic heart disorder, cardiovascular disease, restrictive pulmonary infection, problems with respiration, hypertension, as well as diabetes mellitus (Ahmed et al., 2016a; Alamdar et al., 2017; Attreed et al., 2017; Bhowmick et al., 2018; Chatterjee et al., 2010; Edmunds et al., 2015; Fendorf et al., 2010; Joseph et al., 2015b; Khan et al., 2009; Kumar et al., 2016; Kurzius-Spencer et al., 2017; Liu et al., 2017; Luong et al., 2018; Ng et al., 2003; Rahman et al., 2018a; Saha et al., 1999; Smedley, 2005; Smith et al., 2000; UNICEF, 2007).

The brutality of the hostile health effects of As poisoning depends on the magnitude of As exposure. The health effects are also associated with the chemical type of As, the inherent toxicity, the time of exposure, and the level of the dose (Mateen et al., 2017). A long-term As-exposed condition is the cause of a large number of diseases and adverse health consequences (Guo and Tseng, 2000; Islam et al., 2017e). There is a positive correlation between elevated As in potable water and the incidence of bladder cancer. Islam et al. (2017e) reported that long-term use of As-enriched groundwater (As level 10 µg/L and 50 µg/L) might be the cause of unfavorable health effects. It is estimated that approximately 1, 200,000 cases of hyperpigmentation, 600,000 cases of keratosis, 125, 000 cases of cancers, and surprisingly, 6,000 deaths due to internal cancer occur in Bangladesh per year owing to As contamination of drinking water and the food chain (Ahmed et al., 2017). This estimation is not certain due to data limitations. Methylation is the most important detoxification metabolic process of inorganic As in the human body. The toxicity of As and its impact on public health could be evaluated by using conventional risk estimation processes, for example, identifying the level of risk, evaluating the dose response, estimating exposure, and characterizing risk as well as risk management (Alamdar et al., 2017; Mateen et al., 2017; Yu et al., 2017). Throughout the dose response investigation, Bibi et al. (2008) reported that the As intoxication risk increases with exposure. Several previous studies found a significant positive relationship between the level of As in drinking water and rate of the mortality caused by different types of cancer (Hsueh et al., 2017; Rahman et al., 2015; Saint-Jacques et al., 2018).

The intake level of As with potable water has not yet been clearly determined. However, a maximum intake of 1 µg/L/day of As would lead to cancer (Cheng et al., 2017). In addition, the WHO advised that consumption of 2 µg/L/day As can lead to skin lesions over several years (WHO, 2011). Therefore, the existing level of As risk to humans needs to be assessed on the basis of experimental epidemiological results and investigational data, along with information about the human vulnerability to As in drinking water. A study was conducted in an As-affected region of Bangladesh and reported that the level of chronic disease of the residents of that area was approximately 47%. Moreover, 69% of the dwellers were found to have arsenicosis with malnutrition (Rahman et al., 2018a; Sharma and Flora, 2018). Banerjee et al. (2017) have demonstrated that biomarkers are an early biological consequence of the ingestion of inorganic As and include the blood level of responsive oxidants as well as their antioxidant ability, the genetic issue of provocative molecules, and cytogenetic changes. The level of As in the nails, hair, and urine is a biomarker of a short-term internal dose as well as the impact of long-term internal doses as causes for skin hyperpigmentation along with palmoplantar hyperkeratosis (Hassan et al., 2003). However, exposure to the level of As of more than 0.103 µg/L in the drinking water caused dermatological sickness in As-polluted areas of Bangladesh (Islam et al., 2017b; Rahman et al., 2018a).

5. Studies of groundwater Arsenic

A number of studies have been performed to explore the problems and find solutions related to the high levels of As in the groundwater in Bangladesh. During the last three decades, many researchers have conducted several research studies covering different aspects such as natural (e.g., geological) and anthropogenic (i.e., pumping of water, mining) causes of As occurrence and mobilization in groundwater, cause

and impacts of As in agricultural land and surface water, bad effects of As on human health and the consequences of socioeconomic difficulties for As victims. As affects the human health at the beginning of life even from the pregnancy (Kile et al., 2014; Lee et al., 2017; Mazumdar et al., 2014; Rahman et al., 2011, 2018b). Kile et al. (2016) conducted a study of the As-exposed women from Bangladesh to evaluate the relationship between anemia and As-proactive skin lesions. They found that As proactive skin lesions occurred roughly three times more among those women who had anemia. In addition, pregnant women are mostly exposed to water and food as well as air sources of As because Shih et al. (2017) demonstrated a significant relationship between prenatal As exposure and the risk of stillbirth. They also argued that there is a high risk of child mortality which depends on the prenatal As exposure. As exposure for the newborns can happen due to the As contamination in the breast milk as well. A most recent study (Bassil et al., 2018) on the influence of toxic metal contamination in the breast milk demonstrated that infant intake of Pb, Cd, and As from the breast milk can be the hazardous elements for them. Moreover, As is the cause of the various types of cancers and the influential factor for different chronic (Joseph et al., 2015b; Ourshalimian et al., 2019).

In Bangladesh, the groundwater As contamination creates widespread social troubles for the victims and for the As-affected families (Hadi and Parveen, 2004; Mondal et al., 2017; Mosler et al., 2010; Paul, 2004; Rahman et al., 2018a). Mahmood and Halder (2011) found that As-affected patients face severely pessimistic social negligence, and there is a significant difference in perceptions about the As and the social issues among the As-affected patients and non-affected people. In the society, As-invaded people are banned from the social activities, even often facing rejection from their own family members (Mosler et al., 2010; Shih et al., 2017). Moreover, As-affected unmarried women are incapable of getting married. In some cases, the As-affected housewives are being divorced. As-affected children are not going to school to hide the problem (Alamdar et al., 2017; Kippler et al., 2016; Minatel et al., 2018; Rahman et al., 2018b). However, As contamination in groundwater also creates widespread economic problems for the As victims. For example, Nahar (2009) found that in a rural area in Bangladesh, the As levels of the male patients are found to be higher than the As level of female patients. The occurrence of the arsenicosis problem is more intense among the poor, which is directly associated with the poverty circumstances (Argos et al., 2007; Chowdhury et al., 2006; Garshick et al., 2017; Rahman et al., 2018a).

Moreover, the As contamination problem in Bangladesh reveals a significant association between the poverty and the arsenicosis (Argos et al., 2007). Many of the impoverished people in the countryside are occupied with either agricultural activities or with daily labor. However, if the employer identifies the employees with arsenicosis disease, then the employer does not appoint them. Even after appointment, whether the employers find their sickness, then As-affected workers lose their jobs immediately (Mosler et al., 2010). Ahmad et al. (2007) conducted a study on the socioeconomic problems related to arsenicosis in As-affected regions in Bangladesh, and they revealed that approximately 58.6% of the respondents had arsenicosis patients in their own family and experienced the common economic crisis due to the arsenicosis. Furthermore, they identified that the As victims were losing their work efficiency and facing financial difficulties owing to being dropped from their work. They were even unable to get a new job due to their lower efficiency in work. However, there are some technologies that have been developed in the countryside of Bangladesh based on scientific methods and indigenous knowledge to remove the total As from water (Azamat et al., 2018; De and Maiti, 2011; Deschamps et al., 2005; Guo et al., 2007; Kabir and Chowdhury, 2017; Mukherjee et al., 2007; Wang et al., 2017, 2019; Zhang et al., 2003). Mondal et al. (2017) attempted to assess the social consequences of the As removal technologies for the lives of the As-affected people.

In Bangladesh, the newly formed (Holocene) alluvial as well as deltaic plains are mostly affected by As while the older (Pleistocene)

alluvial is less affected (Nahar, 2009; Reza and Jean, 2012). BGS and DPHE (2001) analyzed the deep groundwater samples collected from coastal areas and recommended that this deep groundwater samples might not be like those somewhere else in Bangladesh. Consequently, they suggested that the quality, quantity as well as sustainability of the groundwater resource of the county need to be estimated along with the consideration of the potential consequences of large-scale artificial groundwater withdrawn for irrigational purposes from the deep aquifers. Rasul et al. (2002) identified the species of As from 18 districts in Bangladesh and showed that 60% of the analyzed samples had the most poisonous As(III) of 10 µg/L, and 44% had 50 µg/L. Specifically, they demonstrated that the As(III) of Bangladesh was distributed within 100–200 feet in the deep aquifers. However, the geochemistry and As distribution of the groundwaters of the Meghna River deltaic plain of Bangladesh was analyzed by Bibi et al. (2008). They also evaluated the As mobilization potentiality from the host sediment into the groundwater and concluded that As is released in the organic fractions of the sediments even though the dissolution of iron and Ca²⁺ is too low. Similarly, von Brömssen et al. (2014) conducted a hydrogeological study in southeastern Bangladesh, and reported that the hydraulic heads are influenced locally and seasonal pumping occurs. Moreover, the groundwater is naturally recharged by monsoonal replenishment. This study identified two-flow systems including: (i) the deeper and regional flow systems that have originated from the recharge regions (i.e., Tripura Hills), and (ii) the shallow and local flow systems originating from the local topography as well as from the local groundwater recharge which increases the level of As in the shallow aquifers. Islam et al. (2017f) argued that the groundwater flow from the Patuakhali district, Bangladesh, is very dynamic, flushing as well as it transports the solute loads to the aquifer with little recharge from the ponds, the rivers and the rice fields. Ponds are receiving household waste water from villages and are acting as another possible source of the organic carbon in the groundwater systems (Alam and McPhedran, 2019; Freikowski et al., 2013; Gong et al., 2015; Whaley-Martin et al., 2017). In addition, irrigation greatly changes the location, time and the chemical quantity of the aquifers. In addition, in the dry season, the large flux cycling of irrigational water throughout the rice fields as well as the biogeochemical cycle of As and Fe (Tareq et al., 2003) could mobilize the As from oxidized near-surface sediments (Harvey et al., 2006). The dissolved organic matter in the groundwater environment of Bangladesh (e.g., Faridpur District, located in the Ganges flood plain and Sonargaon from the Meghna flood plain) exhibits a significant relationship between the fluorescence intensity of the small molecular fraction and the As concentration that plays a vital role in the As and toxicity mobilization (Tareq et al., 2013). A notable work by Hossain et al. (2013) was conducted in the Kushtia district, and the authors mentioned that the reductive solution of MnO(OH) as well as FeO(OH) that is mediated by the anaerobic bacteria subsequently acts as the major mechanism to mobilize As in the groundwater. In that study, they used groundwater samples without analyzing isotope geochemistry. Studies on the presence of As in soil, plants, and vegetables in Bangladesh have also been conducted.

After analyzing groundwater and soil samples, Das et al. (2004) mentioned that all the water and soil samples crossed the accepted limits of As (10 µg/L). At the same time, they showed that the rice grain did not contain As concentrations above the WHO suggested value (10 µg/L) but the roots of the rice plant had elevated As concentration. Moreover, As contents of some vegetables [potatoes (*Solanum tuberosum*), Kachu sak (*Colocasia antiquorum*), as well as Kalmi sak (*Ipomoea reptans*)] exceed the food safety limit. Similarly, Alam et al. (2003) found that some other vegetables such as green papaya, snake gourd, elephant foot, ghotkol, taro, and the leaf of bottle ground also contain elevated levels of As. Some of these vegetables also accumulated some heavy metals (e.g., zinc, lead, and copper). A huge number of the people of Bangladesh take in As via water and food (Bakhat et al., 2017; Islam et al., 2017e; Ohno et al., 2007). This intake is happening due to the irrigation with

As-contaminated groundwater in the agricultural land (Das et al., 2004). As is also polluting the soil of Bangladesh (Islam et al., 2017d; Shrivastava et al., 2017; Van Geen et al., 2006). Irrigation water flow influences the As transportation system into the naked rice-field soils and puts As into the groundwater through abiotic as well as biotic desorption (Polizzotto et al., 2013). Recently, Chowdhury et al. (2017) mentioned that the As concentrations in the paddy soil is higher relative to the non-paddy soils. They also argued that those soils, irrigated with groundwater, had higher As concentrations than the soils irrigated by the surface water. They differentiated the As concentration between the Holocene flood plains and Pleistocene terraces and concluded that the Holocene flood plain soils contain more elevated As than the other elements. In consideration of their study, it is also obvious that As is most probably correlated with the weathered or leached soils owing to the young geological setting of the Holocene sediments.

However, several explorations have been performed to investigate As mobilization mechanisms and the As source(s) in the groundwater of Bangladesh. The investigators have ensured that the dissolved natural matter and the inorganic solution of Fe–Al oxyhydroxide are typically responsible of mobilizing As in the aquifers of Bangladesh (Cui et al., 2018; Martin et al., 2015; Pi et al., 2017b; Radloff et al., 2007). Moreover, reductive suspension of MnOOH as well as FeOOH mediated from anaerobic microbes is considered the major mechanism to release As in the groundwater (Gomez-Gonzalez et al., 2016). In addition, human activities (such as irrigation) also play a significant role in the timing, forms of the chemical and setting of an aquifer (Xiao et al., 2017) even in As mobilization (Herath et al., 2016). However, the potential geochemical factors of As release and hydrogeochemical variation in low and elevated As groundwater are still not clear (Cui et al., 2018; Maier et al., 2017; Xu et al., 2017; Zhang et al., 2017c). Moreover, the geochemical processes of high As groundwater aquifers have not been explored absolutely, and the major origins of As in groundwater are not yet well understood (Abass et al., 2016; Bibi et al., 2017; Hasan et al., 2009; Nazari et al., 2017).

Meanwhile, numerous studies have offered significant information concerning the groundwater geochemistry of Bangladesh. Most of those research studies were conducted by the conventional techniques of the hydrogeological investigations and explored to find the reasons for As enrichment and the factors of As release in groundwater. For instance, Bibi et al. (2008) evaluated the groundwater geochemistry of the Meghna River delta in Bangladesh to show As distribution and identify the potential As mobilization processes in aquifers. Similarly (Chowdhury et al., 2017; Haque et al., 2015, 2017), discussed the causes of As occurrence and geochemical factors controlling the processes of As mobilization in the groundwater of Kushtia District, Bangladesh. However, a remarkable study related to isotopes and hydrochemical tracers was performed by Majumder et al. (2013). They showed the connectivity of the river water and adjacent riverbank aquifers of southern Bangladesh. However, very few studies have focused on the evaluation of the hydrochemical as well as physical processes in the groundwater of Bangladesh using the hydrochemical and environmental isotopic data. To fill this research gap, need to characterize the major hydrogeochemical elements along with environmental isotopes of the groundwater.

Research on the natural occurrence of As and other trace metals (TMs) in the groundwater and their consequences on human health has been conducted in some Asian countries and in Bangladesh as well. For instance, Rahman et al. (2016a) have shown the geochemical control of As occurrence as well as the distribution in the district of Manikganj, Bangladesh. Similarly, Islam et al. (2017c) have revealed the spatial distribution of TMs in the groundwater of Rangpur District, Bangladesh. Previously, Bayatkashkoli et al. (2017) demonstrated that the concentrations of boron (B) and As are high in the floodplain agricultural soil in Bangladesh. However, so far, the concentration of As and B in groundwater of the country is a major issue of concern. These studies focused on the As and TMs distribution in groundwater systems and exhibit a

distinct pattern in various areas in Bangladesh. Nevertheless, the co-occurrence of As and TMs in groundwater and the mechanisms of As release have not been explored in detail.

However, numerous studies (Bhattacharya et al., 2009; Cui et al., 2018; Freikowski et al., 2013; Guo et al., 2014; Postma et al., 2017; Rahman et al., 2016b; Tareq et al., 2013; Xu et al., 2017) have been completed and different hypotheses have been introduced to manifest the roots of elevated As concentration along with As release mechanisms in the groundwater. Tareq et al. (2013) noted that the incidence of As resulted from the geochemical as well as biogeochemical sequences of iron and sulfate. In addition, As mobilization is associated with the scrupulous behavior of natural matter. Some researchers (Duan et al., 2017; Flora, 2015; Jung et al., 2015; Pi et al., 2017b; Schaefer et al., 2017) have confirmed that redox environments are directly or indirectly coupled with the microbial activities as the typical mechanism to control the As mobilization process in aquifers. On the contrary, occurrence of high As in groundwater and As mobilization is associated with geochemically reduced subsurface environments and governed by the native geology and geomorphology as well as hydrogeology (Kinniburgh et al., 2003; Rahman et al., 2016b). In addition, As occurrence in the groundwater of Bangladesh is also linked to the hydrogeochemistry of sediment and groundwater along with anthropogenic activities. Various studies (Bibi et al., 2017; Das et al., 2018; Donselaar et al., 2017; Kinniburgh et al., 2003; Reyes et al., 2015; Reza et al., 2010b; Smedley, 2005; Verma et al., 2015; Welch et al., 2000) have mentioned that the biogenic reduction of Fe-oxyhydroxides with carbon inflow resulting from the large-scale irrigation are the major mechanisms to increase the geogenic As in the groundwater. The groundwater As of Bangladesh is mobilized by a combination of the physical and human processes (for instance, natural weathering, biological factions, and anthropogenic activities).

6. Sources of Arsenic

The Rajmahal Traps along with the Rajmahal Basin from India (adjacent to Bangladesh) are believed to be the potential sources of elevated As minerals (Anawar et al., 2002; Ayers et al., 2016). The sediments from the high As aquifers usually show high As content. Moreover, the hydrologic as well as the biogeochemical circumstances trigger the As mobilization process from these types of sediments under slow flow rate conditions (Freikowski et al., 2013; Xie et al., 2012a). In contrast, Wang et al. (2019) reported that the As-rich aquifers usually are situated adjacent to the young fold in mountainous areas. Hasan et al. (2009) claimed that the high As contaminated regions of the earth are situated in and/or near to the large sedimentary basins neighboring the main orogenic belts, demonstrating the significant role of the regional geological setting in controlling the As concentrations. The sources of As are discussed in the following section.

6.1. Primary sources

6.1.1. Geographical location and climatic condition

The land surface of Bangladesh is developed from the GBM flood plain. The base metal sediments of the GBM flood plain are excessively localized and act as the potential origin of As. Gondwana coal of the Rajmahal contains high As, and the overlying basaltic rock is drained through the Ganges River as well as its tributaries. Thus, this Gondwana coal adds sediment in this area after it acts as the source of As. The tributaries of the Ganges flow contain base-metal deposits from the eastern Himalayas, and it is also believed to be a probable As source. In addition, the alluvial deposits of this area transport As from the Himalayas. Owing to the shallow buried thickness of the superficial groundwater, arid climatic conditions might lead to mobilization of As in groundwater and then enrich the shallow aquifers. In terms of deep confined groundwater, climatic conditions had a slight effect on As enrichment.

6.1.2. Hydrogeological circumstances

The geology of Bangladesh is generally distinguished by the alluvial as well as deltaic sediments developed from the GBM flood plain, which results from the Himalayan weathering (Dowling et al., 2002; Haque et al., 2015). The two main kinds of geological formations (Pleistocene antique alluvium and the Holocene sediments) are common in Bangladesh. This area is largely consisting of two sub-surface beds, including the top clay-silt-sand bed and the sandy bed with different sizes of grains. The formation of a sandy bed can be sub-divided into the fine, medium-fine, medium-coarse, and coarse sand as well as coarse sand along with gravel (Fig. 2). The shell geology is developed mostly from the flood plain. In addition, the surface area is still categorized as a reactive flood plain. This new flood plain includes clay and silt as well as fine-to-medium sized sand which are relatively slack and easily broken in nature, which facilitates the As enrichment process from shallow to deep aquifers (Islam et al., 2015).

The rainwater and surface water (e.g., pond, river) with the atmospheric gas infiltrate into the water-saturated zone. This water then enters the phreatic zone and is isolated from the atmosphere. The aerobic organisms consume oxygen to metabolize the organic matter and introduce carbon dioxide into the groundwater. After using all the oxygen, anaerobic microbes are converted to iron oxy-hydroxides to gain energy that introduces CO₂, Fe, and As into the groundwater. The occurrence of dissolved As is controlled mainly by the biological activities and adsorption reactions (Fig. 3).

The major origins of sediments serve as the most important source of As. The As distribution map of Bangladesh (Fig. 1) shows that elevated As contamination occurs within the catchment area of the GBM river system, which strongly represents the various influential provenances (Edmunds et al., 2015; Jung et al., 2015). It also demonstrates that the Himalayas, situated in the north of Bangladesh, act as the parent rock of As (Hossain et al., 2013; Nath et al., 2008). Based on the previous studies (Ayers et al., 2016; Dowling et al., 2002; Freikowski et al., 2013; Smedley and Kinniburgh, 2002; Whaley-Martin et al., 2017) the potential As sources include ① the coal of Gondwana containing high As of the Rajmahal Trap region and the overlying basaltic rock of its drained through the river of Ganges as well as its tributaries. Thus, this Gondwana coal adds sediment into Bengal Basin and subsequently it acts as the source of As in this basin; ② the tributaries of the Padma river mainly in North Bengal flowed via the Gorubathan base metal deposits carried with As enriched sediments from the eastern part of Himalayas that is also considered as a possible origin of As; ③ the Bihar mica belt

(As concentration ranging from 0.08 to 0.12%) also believed the potential As source; ④ the alluvial sediments transport As from the Himalayas. It is mostly accepted source of As in this area; and ⑤ under the euxinic environment, the prevailing of the paleo-channels of Padma river, As occurs as the biogenic deposit.

Volcanoes as well as fumaroles might function as the key primary As source. However, volcanoes are not supposed to contribute As to the groundwater of Bangladesh (Rahman et al., 2016a). The distinct sources of As in the aquifer of Bangladesh are still in need of exact identification. The Siwalik Cape is mostly formatted by the fluvial greywacke sand-stones with some of the silt as well as the clay. Several hot springs are present in the margin of Siwalik that transport As-contaminated magmatic water into the aquifers. Moreover, the mineralogy along with the fine grain of the Siwalik sediment containing high As can also be a cause of elevated As, which moreover exists in Fe and/or S-bearing minerals (Chakraborti et al., 2015; Wang and Shpeyzer, 2000; Yang et al., 2017). The Himalaya-derived As contaminated sediment of the Siwalik basin is hence defined as the depository source of As in Bangladesh (Fig. 4). Moreover, the ophiolite of the Indus-Tsangpo stitch zone is also regarded as a primary source of As, which was ultimately eroded from the Siwaliks in the period of Miocene as well as Pleistocene and consequently weathered significantly during the Holocene (Huq et al., 2018b; Neidhardt et al., 2018; Radloff et al., 2017; Wang et al., 2019). In addition, the Quamdo-Simao plutonic and ophiolitic region located near the India-Bangladesh border was eroded, then transported with As-enriched deposits to the foreland of the Siwalik basin (Fig. 4), also considered one of the primary sources of As.

The key source minerals of As are sulfides, typically pyrites, and the metamorphic original rocks of the Himalayas (Pi et al., 2017a). Since the late Pleistocene, the weathered oxidative parent minerals directed the release of the secondary phases of iron oxides and clay particles along with hydroxides to river sediments. This geochemical process transferred a large amount of co-weathered As from the mafic and sulfide mineral stages to the recently created secondary phases (Anwar et al., 2002; Hasan et al., 2009; Zhang et al., 2017b). It effectively sorbed a large quantity of As into its shell and then functioned as the main source of As in this area. However, von Brömssen et al. (2014) mentioned that the sediment of Bangladesh contains As carrying pyrite grains. However, the deep Pleistocene sediments have suffered for a long period with oxidative weathering. On the contrary, the shallow Holocene sands were mainly derived in the extended glacial period resulting from physical weathering and act as a principal source of moveable As (Das et al.,

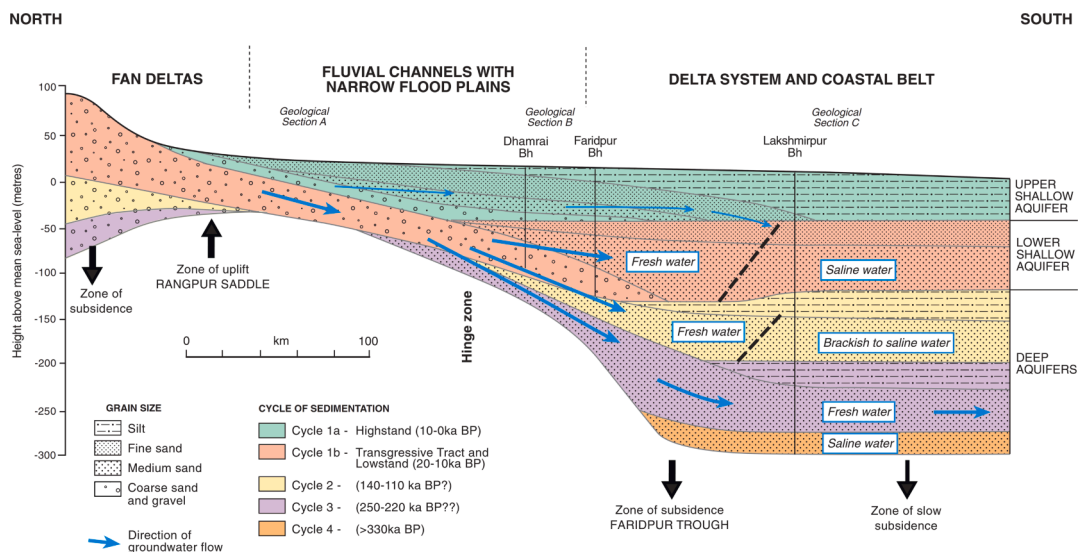


Fig. 2. Hydrogeological cross-section of Bangladesh. Particularly shown are the geological structure and groundwater flow patterns within Mid to Upper Quaternary sediments (BGS and DPHE, 2001).

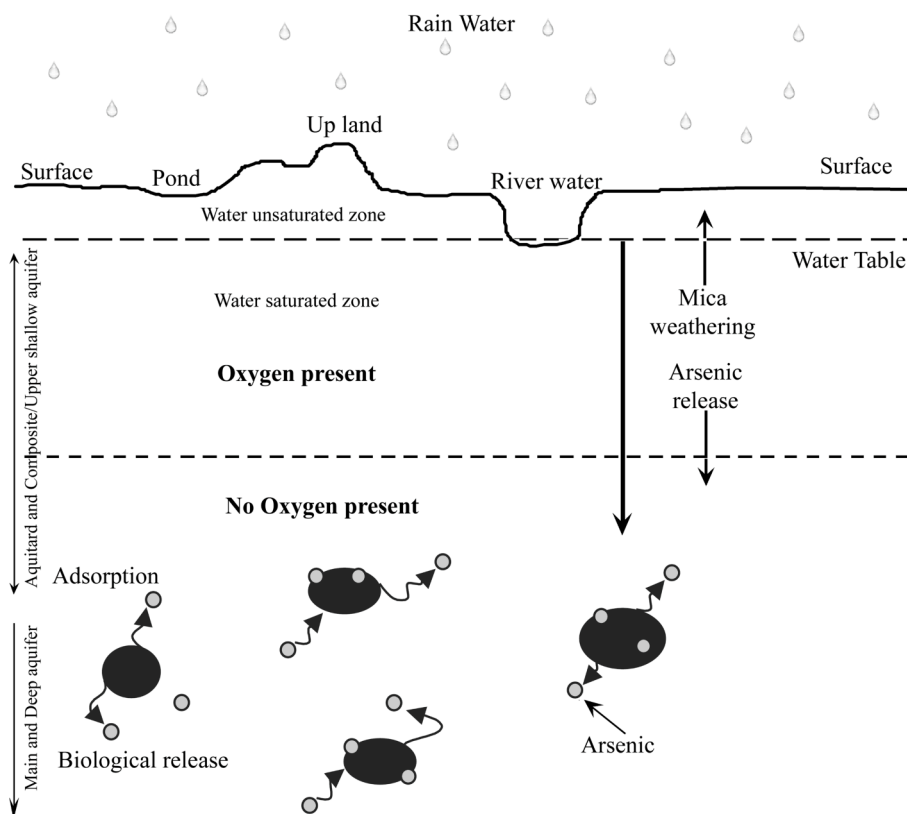


Fig. 3. Schematic diagram of groundwater system of the study area (modified from (Dowling et al., 2002)).

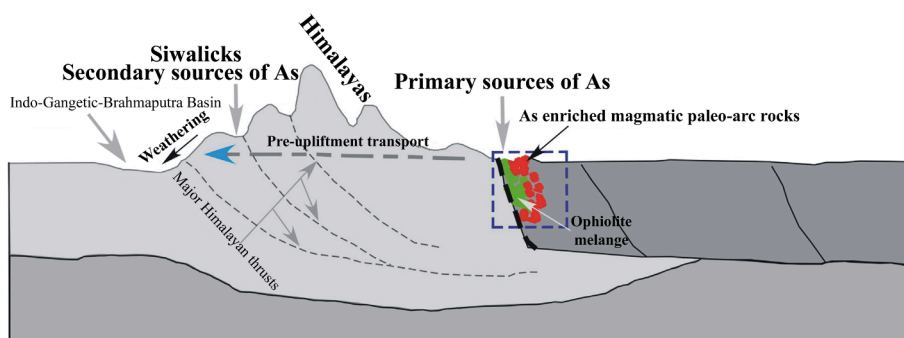


Fig. 4. A hypothetical model of primary and secondary sources of groundwater As (modified from (Mukherjee et al., 2014)).

2018; Xie et al., 2014). Fig. 5 shows the potential processes of As occurrence in the groundwater in Bangladesh.

6.1.3. Runoff conditions

The groundwater flow of Bangladesh is directed from north to south. Therefore, it is difficult for the groundwater As enrichment in deep aquifers due to the favorable runoff conditions. Additionally, the vertical sketch of As concentration (Fig. 6) confirmed that the shallow groundwater is more As concentrated than the deep groundwater. The upper aquifers were relatively loose and very fragile, but the deep aquifers were formed by the fine particles under the poor runoff conditions within an enclosed environment. As a result, the upper (shallow) aquifers accumulated high As enrichment.

6.2. Secondary sources

Mobilization of the little fraction of As is sufficient to increase the As concentration in groundwater higher than its permissible level. The

solid phase of As in the sediments is usually bound to the iron hydroxide as well as the detrital sulfide mineral (Dai et al., 2016; Pi et al., 2017a). Iron enriches mineral stages, namely, oxides, silicates, hydroxides, and carbonates contain considerable amounts of As. This iron acts as the main host of the As in the groundwater of Bangladesh (Kanoua and Merkel, 2017; Radloff et al., 2007; Stollenwerk et al., 2007). Secondary sources such as redox conditions, role of pH, and total dissolve solid (TDS) are the key sources of As in Bangladesh.

6.2.1. Redox environment

Redox conditions of the groundwater are considered the key factor of As dissolution. However, on the basis of the hydrogeochemical environment, high As concentrations could occur in oxygenated and/or in reduced waters (Howladar, 2017; Su et al., 2016). The hydroxides of iron might result from the mica weathering, iron sulfide and different Fe carrying minerals (Liu et al., 2013) that might be adsorbed from the iron hydroxide molecule surface, due to the potent adsorption ability of several iron hydroxides. The adsorption process of As from iron

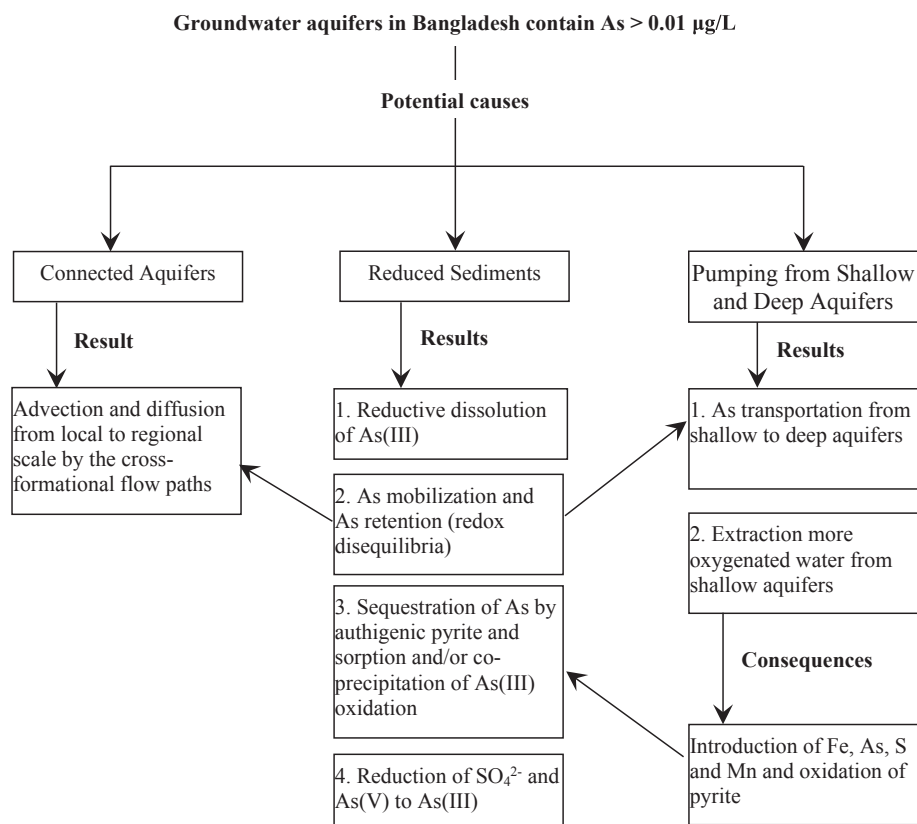


Fig. 5. The potential features and processes of As occurrence in groundwater in Bangladesh (modified from (Mukherjee et al., 2011)).

hydroxides might occur with anion adsorption, in addition uptake of the protons and/or release of the hydroxide ions (Kanoua and Merkel, 2017; Meng et al., 2017; Radloff et al., 2017; Reza and Jean, 2012). The surface loading of iron hydroxides to sediment is more active in the Holocene orange sediment than in the Pleistocene gray sediment, due to the minor level of the oxidative weathering. This iron hydroxide acts as a sorbent to increase the amount of dissolved As in groundwater (Huq et al., 2018b). However, Knappett et al. (2016) questioned the accuracy of the ferric hydroxide since the source of As in the reduced aquifer of Bangladesh because in Holocene aquifers, only ferric hydroxides are regionally limited or absent, as illustrated by the whole surface FeOOH loading in the Holocene aquifers (Radloff et al., 2017).

However, pyrite is very usual sulfide mineral and present in the reduced alluvial aquifers in Bangladesh (Chakraborti et al., 2015; Das et al., 2018; Flora, 2015). Authigenic framboidal as well as huge pyrites could contain up to 11,000 and 13,000 mg/kg of As, respectively, and this distribution can account for approximately 70% of As among the total As of the aquifers in Bangladesh (Lowers et al., 2007). Bhattacharya et al. (2009) argued that the detrital and the grains of authigenic sulfide bear approximately 60% of the As within the total As of the Holocene aquifers of Bangladesh. A number of studies have reported the important role of clays for the sorption of As in aquifer (Bayatkashkoli et al., 2017; McArthur et al., 2001; Reza and Jean, 2012). Organic matter enriching the fine-grained shale and clay is found with highly enriched As. However, several studies have emphasized the importance of iron oxide-coated sand and grains of mica of the sediment as the potential adsorbent of As (Das et al., 2018; Freikowski et al., 2013; Tareq et al., 2003).

6.2.2. pH, TDS and F⁻

The level of pH in groundwater is a vital factor for As enrichment in groundwater (Wang et al., 2019). The groundwater of Bangladesh is weakly alkaline in nature (Huq et al., 2018). Alkaline conditions may

also contribute to the desorption of As as well as enhance groundwater As. However, As could easily be adsorbed by positively charged minerals, namely, Fe or Al oxides. With increasing pH values, the positive charges are carried by the colloids as well as clay minerals. Huq et al. (2018b) observed that the elevated As in groundwater was accompanied by elevated pH values that ranged between 7.65 and 8.3. In addition, they found a positive relationship (0.464) between the As concentration and TDS. However, the elevated As groundwater had a low level of TDS, indicating that TDS had small control over As enrichment in groundwater. Huq et al. (2018b) mentioned that there was no significant correlation between As and F⁻ concentration among the samples analyzed and demonstrated that F⁻ had either no or less control over As enrichment in the groundwater of Bangladesh.

6.3. Tertiary sources

The As in soils in Bangladesh is observed at levels up to 40 mg/kg on the surface. This high occurrence of As in the soil might be due to irrigation runoff (Polizzotto et al., 2013). However, Chowdhury et al. (2018) mentioned that As in soil was at an elevated level, even earlier than the start of irrigational pumping. Rahman et al. (2015) noted that irrigated soil obviously has a higher As concentration than non-irrigated soil. In Bangladesh, widespread irrigational pumping has been practiced at a large scale from the beginning of the 1970s (Fransisca et al., 2015; Mukherjee et al., 2017). Irrigational pumping brings anoxic groundwater from the aquifer into contact with atmospheric oxygen. Subsequently, the As withdrawn due to irrigational pumping is seized by an oxidized ferric iron in the farming fields. As a result, the widespread rice fields of Bangladesh increase in As content by approximately 1 kg/ha/year in the soil zone (Polizzotto et al., 2013). Therefore, this As might then be recirculated in groundwater, causing As contamination in aquifers. Specifically, contamination occurs in the dry season. Islam et al. (2017f), Polizzotto et al. (2013), and Rahman et al. (2018a)

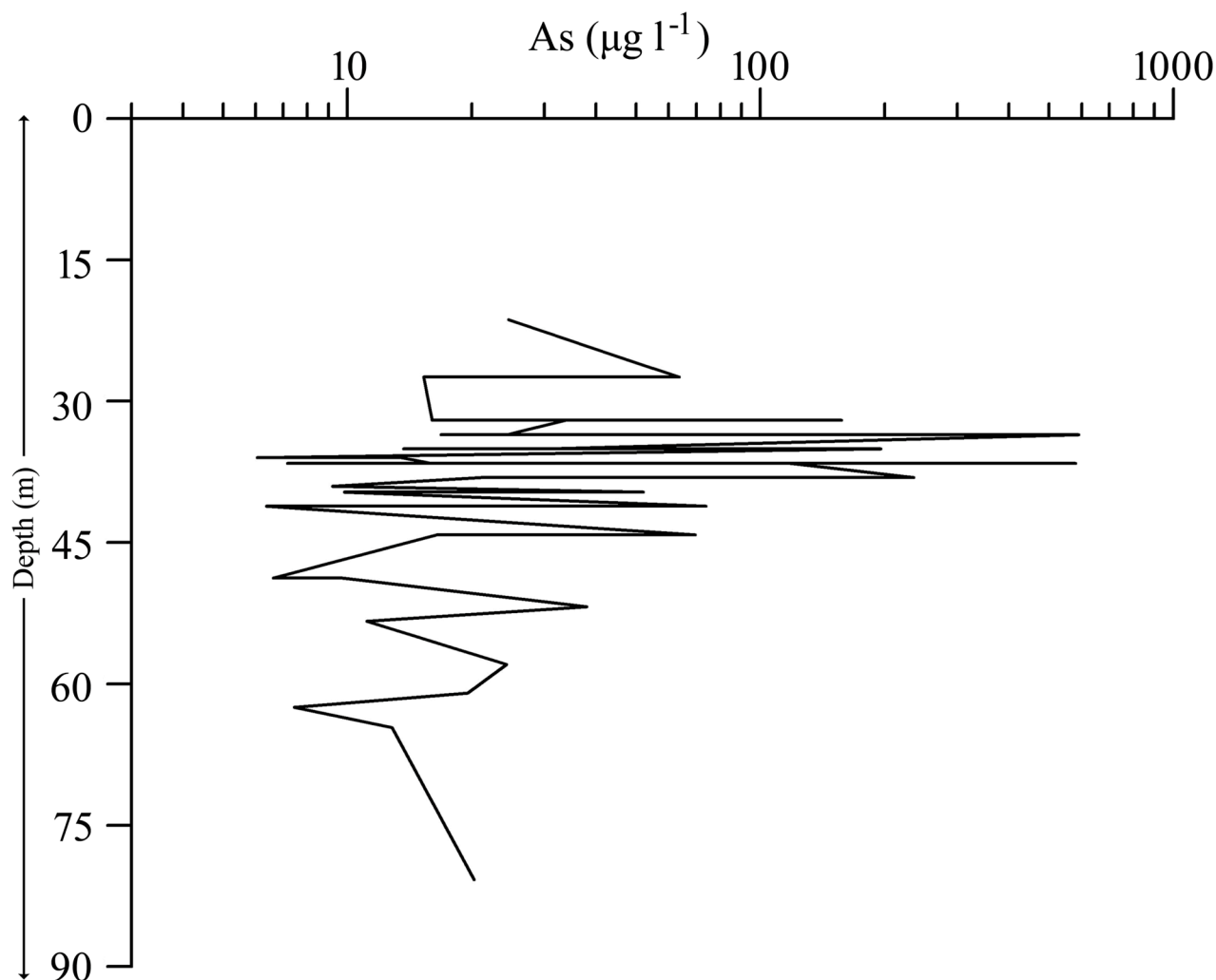


Fig. 6. Correlation plot of As concentration in groundwater with the depth of wells (Huq et al., 2018b).

indicated that approximately 1000–1360 tons of As is added every year to the soil of Bangladesh. Recharging water from the irrigational return flow is probably enriched in natural matter and naturally anoxic as well, so the return flow influences the reduction process of metal hydroxide on the surface and in the deep aquifers.

Polizzotto et al. (2013) argued that not only does the irrigational return flow add such high levels of As to the surface layer but the newly deposited sediment also contributes As to the soil. The authors hence reported that the elevated As concentrations in soils are due to the use of groundwater for irrigational purposes, and the permanent inflow of As-laden sediments onto surface soil. They mentioned that the surficial sediment is the leading source of As in groundwater that undergoes cycling of seasonal redox and results from the mobilization process. During the monsoon flooding, the soil releases a huge amount of As into the floodwater, accounting for 13–62% of the overall amount of As that is added each year to the soils of Bangladesh. This As subsequently moves across the rivers as the floodwaters recede (Roberts et al., 2010). Yu et al. (2015) demonstrated that, between pre-flood and post-flood, the occurrence of the average As content in the shallow groundwater of Datong Basin, China, is increased from 22.6 to 31.1 µg/L. They also observed that a minor change occurred in the As concentration (from 14.0 to 5.1 µg/L) in surface water, indicating that the return flow of As-enriched floodwater added extra As to the groundwater. Finally, they concluded that the As concentration of the groundwater samples obviously increased after a flood event. The increase of As in the groundwater after flooding could not occur only by mixing. The increase of As

in the groundwater also resulted from the additional As mobilization from aquifers. Flooding is also responsible for changing of the geochemistry of high As groundwater (Roberts et al., 2010). Moreover, runoff from the human activities is also responsible for contaminating the surface water as well as the groundwater. In the deep subsurface, an elevated As concentration is associated with the compaction caused by groundwater withdrawals. However, the primary risk of As contamination is increased, which could also cause intense secondary changes in the geochemical environment. Furthermore, As also increases due to mining activities (Bose and De, 2013).

7. As mobilization

To date, it is obvious that both natural and the anthropogenic practices are responsible for enhancing As in groundwater (Kinniburgh et al., 2003; Reyes et al., 2015; Reza et al., 2010b). The high As intensity is associated with rock weathering, natural floods, volcanic eruptions, bushfires, and leakage in the geological formations of the Himalayan ranges. Anthropogenic activities such as mining, use of pesticides and fertilizers on farm land, and chemical industries allow As into the environment (Bibi et al., 2017; Kinniburgh et al., 2003). Irrigation with elevated As-contaminated groundwater in arid and semi-dry regions also intensifies the As concentration in the alluvial aquifers (Tanvir Rahman et al., 2017). Many recent studies (Diwakar et al., 2015; Maier et al., 2017; Pi et al., 2017b) have attempted to understand the mechanisms of As mobilization in the groundwater. Different existing theories

as well as competing theories such as pyrite oxidation (Lowers et al., 2007), competitive exchange (Hasan et al., 2009), and the dissolution of the iron oxy-hydroxides (Xu et al., 2017) have already been introduced to explain the As release process in groundwater. However, various processes may be activated to mobilize the As in the groundwater in Bangladesh (Fig. 5).

7.1. Oxidation reduction

Reductive dissolution of the iron hydroxide derived from the degradation of microbial mediation of the organic carbon of sediments that release the sorbed As into the groundwater is the most accepted hypothesis to explain As mobilization in the groundwater environment (Nazari et al., 2017; Radloff et al., 2007). Xu et al. (2017) stated that the reaction that controls the reduction process as well as iron (III) hydroxide dissolution to Fe^{2+} by generating HCO_3^- with liberation of whole sorbed load to the iron hydroxides (including As) is responsible for mobilization of As in the groundwater. This process reveals the significant correlation between As and HCO_3^- (Gao et al., 2013). Ahmed et al. (2004) argued that decomposed organic matter adds an elevated level of dissolved natural carbon to the groundwater. Moreover, the authors mentioned that oxidative decomposing organic matter could raise the concentration of HCO_3^- . The additional HCO_3^- might be supplied into the groundwater aquifers by the local biogeochemical action (Zhang et al., 2017a). Harvey et al. (2006) noticed that the reduction processes in groundwater along with the weak but positive relationship of the dissolved As to the HCO_3^- in the groundwater of Bangladesh indicates that the As may be mobilized with the reductive dissolution and reductive desorption from the arsenate to arsenite. In addition, the elevated As concentration in the Holocene aquifers could contribute by completing the dissolution of the FeOOH driven from microbial metabolism of the organic matter (Knappett et al., 2016; Radloff et al., 2017).

Chen et al. (2017) claimed that the microbial activity might mobilize As in the groundwater. However, Bhattacharya et al. (2009) argued that the biogeochemically catalyzed cycling of Fe(II)–Fe(III) in the aquifer probably acts as the key process to control the As mobilization. They also mentioned that the cyclic attitude of Fe might also be attributed to the existence of nitrate in the groundwater that is mainly supplied from the anthropogenic sources. Dai et al. (2016) argued that the mobilization of As mostly results from the microbial reduction of (Fe(III)/Mn) hydroxides. Smedley and Kinniburgh (2002) demonstrated that the desorption along with the reductive dissolution of the Mn oxides might also contribute to the groundwater As concentration like FeOOH. Tareq et al. (2013) reported that in Bangladesh, As is present in the biotite. Similarly, Xie et al. (2014) argued further that even though the hypothesis of iron hydroxide dissolution is very widely accepted, biotite weathering could also add significant As into the groundwater.

7.2. Reduction in As species

The microbial deterioration of the organic matter acts to reduce Fe(III)–Fe(II) as well as assisting in the reduction of As(V) to As(III) (Postma et al., 2017). The reduction system of arsenate to arsenite, led by the mediated microbial process, is believed to be responsible for the mobilization of As in the groundwater within the strongly reductive circumstances of the shallow aquifers. The As(III) in the shallow aquifer systems in Bangladesh not only causes the redox conditions of the aquifer but also contributes by the microbial reduction of As(V) (Nazari et al., 2017). However, the anoxic condition promotes the dissimilar reduction of As(V), whereas the anaerobic microorganisms obtain energy from the combined oxidation of the organic matter. Under these conditions, the decrease of As(V) functions as the marginal electron acceptor for microorganisms (Freikowski et al., 2013; Hasan et al., 2009). Moreover, the dissimilar microbial declination of the As(V)–As(III) could also be moved with the existence of compatible substrates, most probably from the similar group of microorganisms that are also

accountable for the sulfate reduction (Liu et al., 2018). However, this kind of hypothesis is questioned by the observation of iron hydroxide because the iron hydroxide can significantly sorb both the As(V) and As(III) under the extensive diversity of the redox conditions (Pi et al., 2017b). However, Deschamps et al. (2005) noted that the Fe(III) reduces bacteria in the sediment from the bioavailable Fe(III) ion as the electron acceptor. In addition, then Fe(III) reduces the active parts of the sediments surrounded by As(V) for the respiration and then mobilize As(III) into the groundwater (Fig. 7).

7.3. Role of organic matter

Tareq et al. (2013) argued that the irrigational pumping introduced inflow with the fresh inorganic carbon from the rice fields. In contrast, organic-rich ponds as well as the bottoms of rivers release the old organic carbon that influences biogeochemical processes to mobilize As, along with natural carbon-induced reduction and replacement of bicarbonate (Gao et al., 2013). The anthropogenic sources such as human wastes as well as the agricultural activities that infiltrate the irrigational return flow from the agricultural fields as well as the surface water sources are considered as the other sources of the organic matter in the groundwater. Due to the increasing rate of irrigational pumping, the natural organic matter (NOM) flow goes down rapidly, and then it mobilizes the As (Reza et al., 2010a; Tareq et al., 2013; Xie et al., 2012a). Moreover, a long period (thousands of years) is required for the pond-induced organic matter to reach the shallow aquifer. This time duration is much greater than that of the age (>50 years) of As-enriched shallow groundwater. Hence, the in-situ organic matter is considered a possible source of the microbially mediated reductive solution of the Fe-oxyhydroxides which consequently mobilizes the As (Hasan et al., 2009; Zhang et al., 2017a). Herath et al. (2016) claimed that surface-derived organic matter could be the geochemical factor to mobilize the As into the shallower aquifer due to the huge population density and the intensive land use pattern. Chatterjee et al. (2010) noted the use of the native cow dung (which is easily degradable and can be decomposed by the microbial action) as a natural cementing component during tube-well construction. The native cow dung might also function as the carbon source that causes the elevated As concentration in the newly installed tube-wells (Huq et al., 2019).

The organic matter of sediments is an important concern since this

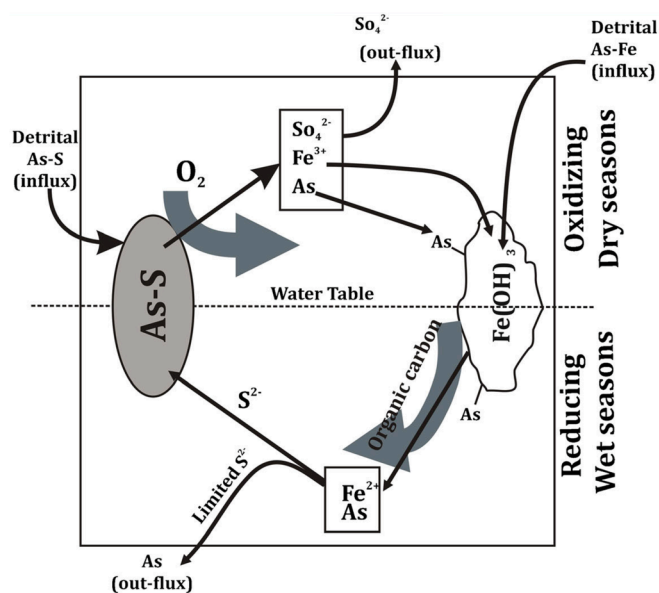


Fig. 7. A theoretical model exhibiting the seasonal variation of oxidation condition of As carrying SO_4^{2-} with formation of Fe^{2+} and As release in the groundwater (adapted from (Polizzotto et al., 2006)).

organic matter is the main source of electron donors. The organic matter of sediments controls the reduction of geochemical processes that can mobilize As (Postma et al., 2017; Xu et al., 2017). In addition, elevated levels of As in As-enriched groundwater are mostly frothy due to the humic acid, which specifies that the high As groundwater may hold elevated levels of natural matter. Thus, it is supposed that the organic matter is a key contributor to create the reductive environment in the aquifer system. The natural matter of groundwater may be attributed to anthropogenic sources such as extensive agricultural activity, using fertilizer and excessive application of pesticides on farmland (Dousova et al., 2016; Shrivastava et al., 2017). Vertical recharge from the irrigation return flow could modify the variation in Cl^- concentration. Additionally, the ponds of Bangladesh have been dug well in the past 50 years. Presently, these ponds are providing a large amount of water for recharging the groundwater. These ponds also receive much household sewage as well as animal effluents. Moreover, in Bangladesh, discharge canals and the ponds are being used for dumping of human as well as animal sewage, putting a large suspended load of natural matter into the groundwater.

7.4. Pyrite oxidation

The model of the pyrite oxidation hypothesis refers to the release of As by oxidized pyrite or arseno-pyrite, most likely owing to the lower water table. This release of As might also result from the large-scale groundwater abstraction from the aquifer systems of Bangladesh (Lowers et al., 2007). The shallow water table represents the anoxic gray sediment of the Holocene that causes the oxidation of the pyrite grains by the soluble sulfate. Then, it releases As into the groundwater. Nazari et al. (2017) also reported that due to irrigational pumping, reduction and oxidation in groundwater have occurred. However, the lower contents of SO_4^{2-} in groundwater is coupled with reducing conditions, and it is believed that As could mobilize from the sulfide minerals (Freikowski et al., 2013). Radloff et al. (2017) reported that the low sulfate is observed in the pores of Holocene aquifers in Bangladesh. In addition, the existence of pyrite particles in aquifer systems suggests that Holocene aquifers are oxidized enough to mobilize As from pyrite. It also acts as the As sink in the groundwater (Lowers et al., 2007; Mango and Ryan, 2015). The hypothesis of pyrite oxidation may not be applicable for the reductive gray aquifers in Bangladesh. If the As is even mobilized from the pyrite oxidation, then it should be reabsorbed from oxidized FeOOH before release into aquifers. Moreover, SO_4^{2-} and As concentration in groundwater is mutually exclusive, so that is incompatible with the As and pyrite oxidation (Bhattacharya et al., 2009; Tareq et al., 2013).

7.5. Role of the ion exchange process

The As species are usually mobilized from the dissolution of its host minerals (Duncan et al., 2017; Mass et al., 2001; Yaghi and Hartikainen, 2018). The ions naturally compete reciprocally, and the adsorbed As species are released to the groundwater (Herath et al., 2016). Silicates might also be the probable competitor of As for the sites of sorption (Chanpiwat et al., 2011; Milošević and Simić, 2015). Cl^- enrichment from the wastewater and the pit latrine is also believed to be the potential species in ion exchange (Caporale et al., 2018; Mukhopadhyay et al., 2017; Nazari et al., 2017; Yang et al., 2017). The CO_2 and HCO_3^- displace the As from the sediment (Gao et al., 2013; Michael and Khan, 2016). The As-enriched sediments along with low concentration of CO_2 are ultimately buried and consequently come into contact with the HCO_3^- -enriched groundwater (Bodrud-Doza et al., 2016). Subsequently, sorbed As is released owing to the competitive extraction of As through the carbonates (Chakraborty et al., 2015; Gao et al., 2013). The most important chemical genus involving in the ion exchange system is the phosphate that may have originated from fertilizer (Mandal et al., 2017; Yang et al., 2017). Moreover, phosphate is supposed to be the key potent competitor in ion exchange with As since silica has weak control in such

exchanging processes but HCO_3^- ions play an important role in the mobilization of As (Aziz et al., 2017; Gao et al., 2013; Zhang et al., 2017c).

The reduction of SO_4^{2-} is a key biogeochemical process that resists the As reduction process in the groundwater environment. The presence of SO_4^{2-} in the groundwater in Bangladesh that has elevated As is usually less relative to the detection level ($<0.01 \mu\text{g/L}$) (Huq et al., 2018b). The concentration of NO_3^- in groundwater is also quite low because of biogeochemical reactions. Thus, low contents of SO_4^{2-} as well as NO_3^- demonstrate that anaerobic bacteria are active in the groundwater system. In alluvial aquifers, Fe(III) , SO_4^{2-} and NO_3^- are the primary electron acceptors for oxidizing organic matter in the groundwater environment. The electron acceptors SO_4^{2-} as well as NO_3^- may be used by the internal microorganisms to produce HCO_3^- from oxidized organic matter (Freikowski et al., 2013; Liu et al., 2018), also indicating that SO_4^{2-} , NO_3^- , and different electron acceptors interact in groundwater and could support the oxidation process of natural matter (Bahar et al., 2016; Mukherjee et al., 2007; Wang and Jiao, 2014) and play a vital role in generating the anoxic environment in groundwater system with elevated As.

The elevated content of HCO_3^- and lower content of SO_4^{2-} and NO_3^- (Huq et al., 2018) obviously illustrate the reductive conditions in the aquifers of Bangladesh that might be associated with the anthropogenic activities (Bose and De, 2013; Zhang et al., 2015). In addition, this high level of HCO_3^- is an indication of microbial functions in the groundwater system. Other studies (Bahar et al., 2016; Mukherjee et al., 2007; Tapase and Kodam, 2018; Wang and Jiao, 2014) have demonstrated that the sulfide oxidation is a major factor in the mobilization of As. However (Huq et al., 2018b), confirmed that the elevated As contamination in the groundwater system in southwestern Bangladesh usually occurs with a low content of SO_4^{2-} . Moreover, SO_4^{2-} concentrations have not exhibited any positive correlation with the As concentration in the different regions of Bangladesh (Huq et al., 2018; Nazari et al., 2017).

The incidence of elevated bicarbonate in the groundwater in Bangladesh might be attributed to the strong biodegradation of organic matter, especially in shallow groundwater (Bhattacharya et al., 2009). Alkalinity has an important function in the hydrochemical development as well as trace metal discharge in groundwater (Alamdar et al., 2017). Earlier studies (Chen et al., 2017; Lin et al., 2017; Maier et al., 2017) noted that the negative relationship between HCO_3^- and SO_4^{2-} can accelerate the As mobilization process. However, others (Lowers et al., 2007; Mukherjee and Fryar, 2008; Mukherjee et al., 2011) argued that the elevated HCO_3^- may be responsible for the release of As in the groundwater of the Bengal Basin. This release of As might result from the completion of HCO_3^- , the chemical inclusion of As and the indefinite electrostatic enrichment of HCO_3^- . In addition, the reduction of Fe would occur with the HCO_3^- to create the pyrite and the siderite precipitates (Kulkarni et al., 2018). However, the occurrence of elevated HCO_3^- concentrations would also lead to the siderite precipitation. We can conclude that the reduction of Fe oxide minerals is a key mechanism in the genesis of elevated As in the groundwater in reducing aquifers.

Different theories have already been developed to illustrate the As mobilization and transportation process in groundwater (Pi et al., 2015). However, these theories do not explain the fact clearly. Harvey et al. (2006) noted that the negative correlation between As and SO_4^{2-} is observed in the groundwater in Bangladesh and demonstrated that As is released from the SO_4^{2-} minerals under reducing conditions. The As in SO_4^{2-} minerals is easily extractable and weakly sorbed. This sorbed As in SO_4^{2-} minerals might mobilize under oxidizing conditions (Ramos et al., 2014). The existence of SO_4^{2-} in the hand pump tube-well water may be contributed by the pit latrines. Groundwater As is also released by the competitive ion exchange of NO_3^- , which is contributed by the fertilizer. In addition, NO_3^- is supposed to be a major key competitor in ion exchange with As. However, as HCO_3^- ion displaces As from the sediments, the HCO_3^- ion plays a significant role in the mobilization of As. Moreover, the As-enriched water containing high HCO_3^- releases sorbed As due to the competitive displacement of As (Michael and Khan, 2016).

The strong positive correlation of Mg^{2+} and Na^+ (Huq et al., 2018b) suggests that Mg^{2+} and Na^+ may result from silicate weathering and/or carbonate suspension and dissolution of the soil salts. Moreover, groundwater with excessive exchangeable Mg^{2+} hinders the infiltration process (Pi et al., 2016). Furthermore, the As contaminated water rinsed out from the soil surface might easily contaminate potable groundwater. However, Chakraborty et al. (2015) reported that the sulfides (generally pyrites) and the metamorphic rocks of the Himalayas are the original mineralogical sources of As. In the late Pleistocene, a huge amount of co-weathered As from sulfide minerals was released to the recently formed Holocene aquifer, which practically sorbs much of the As to the surface and then acts as the major source of As (Lin et al., 2017).

The previous studies also demonstrated that during the cation exchange process, As may be desorbed from the clay minerals in significant quantities and then enters the aqueous phase (Bahar et al., 2016; Su et al., 2016). Several studies (Martin et al., 2015; Postma et al., 2017; Reza et al., 2010a; van Geen et al., 2014; Yaghi and Hartikainen, 2018) also suggested that the Ca^{2+} can increase the As adsorption on Fe oxyhydroxide considerably, particularly at elevated pH. In contrast, Martin et al. (2015) have mentioned that the calcium or magnesium adsorption along with the HCO_3^- could intensify the As desorption from the ferrihydrite to a greater extent relative to their individual competition. However, the existence of HCO_3^- could change the surface charge characteristics of Fe oxyhydroxide through the internal monodentate mononuclear complication and voluntarily compete for adsorption onto Fe oxyhydroxide, resulting in the As desorption. However, competitive adsorption or desorption reactions within the aquifer plays a significant role in the process of mobilizing As. For the deficiency of probable competition in adsorption areas, the As release may be induced by Fe-oxyhydroxide reduction, which can be re-adsorbed to the other Fe phases as well as other sorbents existing in the aquifer. It appeared that the reduction of Fe-oxyhydroxide was pursued with the competitive ion adsorption. In addition, the sediment of the aquifer is responsible for enriching the As in the alluvial aquifers of Bangladesh.

7.6. Microbial processes of As mobilization

According to the different studies (Duan et al., 2017; Huq et al., 2018; Jung et al., 2015; Pi et al., 2017), redox reactions are directly or indirectly associated with microbial behavior and a major mechanism to control the release of As in the groundwater system. In Bangladesh, the occurrence of As and As mobilization are coupled with subsurface geochemical reduction and biogenic reduction of Fe (hydr)oxide that puts As into sedimentary aquifers. Sulfate reductive bacteria play a key role in SO_4^{2-} reduction (Freikowski et al., 2013). Acinetobacter as well as Brevundimonas are very well known bacteria that play a significant role in the As carrying process (Guo et al., 2015; Hare et al., 2017; Liu et al., 2018). Low SO_4^{2-} content reveals the high reduction ratio of SO_4^{2-} , which shows the influence of the sulfate reductive bacterial genus in the groundwater environment. This type of biochemical process increases As incidence in the groundwater in Bangladesh. Moreover, the staid character of Cl^- and the decreasing level of SO_4^{2-}/Cl^- ratios demonstrated the removal of sulfate from the groundwater, indicating the bacterial reduction of SO_4^{2-} in the aquifer system of Bangladesh. However, in the groundwater aquifer, the As concentration is increased with the decrease in the SO_4^{2-} concentration (Huq et al., 2018b), demonstrating that the As is released in the reducing environment together with the reduction of SO_4^{2-} . Furthermore, sulfate-reducing bacteria reduce both NO_3^- and Fe, which would mediate the As mobilization process in the groundwater. Additionally, Fe or Mn oxide reduction continues or is performed before SO_4^{2-} reduction (Guo et al., 2017). As is mobilized in a reductive environment of Fe, Mn, SO_4^{2-} , and NO_3^- (Islam et al., 2004). Therefore, microbe-associated biogeochemical processes play a significant role in the mobilization of As in the groundwater of Bangladesh.

8. Existing policies and guidelines to address the As issue

Since As in the groundwater in Bangladesh has been documented as a threat to human life, considerable efforts have been made towards confirming the presence of safe potable water either by mitigation techniques or discovering of alternate water sources (Huq et al., 2003). Different mitigation actions have been initiated by several organizations, but most these actions have not reached their projected outcomes owing to the technical and spatial as well as socioeconomic challenges (Goel et al., 2019). In 1998, the first National Policy for Safe Water Supply and Sanitation (NPSWSS), a most vital policy, aiming to provide a safe rural water supply with complete coverage in the urban areas was introduced. The policy did not address the quality issues, budget and time frame for achievement. However, in 2004, the National Policy for Arsenic Mitigation and Implementation Plan (NAMIP) was specifically formulated to solve the groundwater As problem. NAMIP offered guidelines for As mitigation in potable water, health, and agricultural purposes. Additionally, the formation of an As mitigation fund was mentioned in this policy (World Bank, 2018). The target of that policy was to ensure safe water (As-free) for drinking, cooking, and agricultural sectors and to diagnose all the cases of arsenicosis as well as managing these cases. NAMIP proposed guidance to mitigate the adverse effects of As on people along with environment in a practical and sustainable way. This policy emphasized the preference of surface water rather than groundwater as a water supply source. However, the policy did not consider problems related to hygiene of water (Mahmud and Mbuya, 2015). Under these national policies, Bangladesh has set and rolled some important sector strategies in the passage of time that covered the Pro-Poor Strategy for the Water and Sanitation Sector (PPSWSS), initiated in 2005 (World Bank, 2018). The PPSWSS 2005, formulated by LGD, was a 10-year scheme for the water supply as well as sanitation. PPSWSS 2005 formed the Arsenic Policy Support Unit (APSU) and the Implementation Plan for Arsenic Mitigation (IPAM) that were guided by the national committee. Nevertheless, it was not continued and in 2008 the APSU had separated, but by 2009 the policy was reformed, and another new initiative was taken to recover the IPAM. The policy recommended taking different approaches to supply As-safe domestic water in accordance with the local circumstances. The key approaches include awareness rising in the rural areas; providing As-removal appliances for the household as well as the community level; deep hand-pump tube-well installation and a construction scheme for a piped water-supply in urban and rural areas as As-safe sources (Tuinhof and Kemper, 2010). IPAM was revised as the Sector Development Plan (SDP) in 2011 as SDP 2011–25 and now includes a joint policy for the water and sanitation sector emphasizing national policies covering the focal areas of concern (confirmation of a safe water supply, mitigation of As contamination). Later, the government disseminated the Water Act of 2013, expected to coordinate, distribute, develop, manage, protect, extract, use, and protect the water resources and conveying a high priority on drinking water (Chan et al., 2016). Another policy was initiated in 2014 with the National Strategy for Water Supply and Sanitation (NSWSS) to explain the objectives and directions of the SDP 2011 and to give a uniform skillful guideline to different sector stakeholders to achieve the safe as well as sustainable water supply (World Bank, 2018). After 2014, the activities, policies and guidelines of the Bangladesh government related to arsenic-free water supply are listed in Table 1.

There are some technologies of As removal from groundwater that also exist in Bangladesh. Those technologies include BUET activated alumina (BUET-AA), Alcan enhanced activated alumina (Alcan), passive sedimentation, Adarsha filter (AR), Stevens Institute Technology (Stevens), DPHE/Danida 2 bucket system, Sono 3-kolshi filter, GARNET homemade emergency filter, and Tetrahedron. Poor performance of the Sono 3-kolshi, Alcan and Tetrahedron with regard to As and/or microbiological pollution in water were experienced in the community-based projects (Goel et al., 2019). However, extensive suitable technologies need to be developed and promoted, and the issues with the prevailing

Table 1
Implementation plan of the national policy and strategy of Bangladesh for water supply and As mitigation.

Time frame	Policy and activities
June 2015	Formulation of exclusive execution plan for As mitigation.
December 2015	Protocol of construction, water quality verification and hand pump preservation or manufacture of wells by the public organizations, NGOs and private sectors. Research associated with the reduction of the As standard of 50 µg/L to 10 µg/L.
June 2016	Updating the database of current As screening in highly affected areas. Proposal regulating the installation of hand pump wells in As-affected areas.
December 2016	Development Project Proposal (DPP) to cover all possible As contaminated areas. Concept note on implications for reducing the As standard.
June 2017	Guidelines on use of surface water as a water supply as well as artificial recharging of groundwater. Concept note on reorganizing and capacity building.
December 2017	Minimum 50 percent As-free pipe water supply. Spread DPP in all remote areas and vulnerable people or prepare a new DPP with these issues by the sector agencies.
December 2018	Master Plans on fresh water supply in urban areas.

millions of As-polluted tube-wells also need to be considered. Research and advanced development of scientific technologies are suggested for the effective recognition of the Millennium Development Goal for safe water.

9. Conclusions

Elevated As in groundwater has been confirmed to be toxic to human health. The As concentration in the groundwater of Bangladesh reaches up to 4600 µg/L. Biotitic chemical weathering and the existence of other core minerals in aquifers are the primary sources of As in the groundwater of Bangladesh. Silicate weathering and degradation of organic matter are very active, which dominate the geochemical process and transports As into shallow aquifers. Groundwater usually occurs in Quaternary alluvial aquifers, which includes shallow semiconfined as well as confined aquifers. Shallow groundwater of sedimentary lacustrine aquifers normally contains high As concentrations, and deeper than 300 m is considered a safe zone. Deep aquifers contain low dissolved As but are in a vulnerable condition owing to the cross contamination from the top shallow aquifers. Large-scale groundwater exploitation and installation of high capacity irrigation wells in deep aquifers could increase the risk of cross contamination. In addition, little infiltration of aquifer sediment, horizontal landscaping, and a slow flow water rate in aquifers are also the result of As. As results from up-gradient sources, mostly from the Himalayas along with drainage through the GBM river channel. Aquifer matrices are considered a direct source of As, which mobilize As into aquifers under reductive conditions. Fe-hydroxides and pyrites are believed to be the most significant host minerals of As. Another probable origin of As is the irrigational recirculation flow from farming lands. All of these sources mobilize As in groundwater under reductive conditions. The elevated As concentration is the consequences of the redox process, microbe reduction, and desorption processes occurring in aquifer systems. Enriched industrial discharge/runoff includes additional As. However, under reductive conditions, both conducive solutions of Fe-oxides as well as reductive desorption are the causes of As occurrence in the groundwater system, leading to As release.

Microbially mediated conducive dissolution of Fe-hydroxides mobilizes As in groundwater. In the reducing environment, microbes, reductive dissolution of Fe-oxides, As(V), pyrite oxidation, and emulsion of an ion exchange process along with redox cycling of surficial soils are also considered to be the probable mechanisms of As mobilization.

Degradation of natural carbon throughout microbe metabolism mobilizes low-molecular weighted natural colloid-bound As and increases the HCO₃⁻ concentration in groundwater. In addition, elevated pH and HCO₃⁻ cause As desorption from the adsorption zones of the aquifers. The high pH and HCO₃⁻ concentrations may be related to microbially mediated Fe(III) reduction, and the oxidative erosion of natural matter creates reductive conditions to mobilize As. However, penetration of the irrigational return flow is also a leading mechanism for As remobilization. Residents should consider using surface water (e.g., pond or river water) as low As concentrated water. This study found the following geochemical processes in the groundwater environment in Bangladesh: (i) chemical weathering, (ii) very active As mobilization, (iii) a competitive ion exchange process releasing As, and (iv) oxidation of sulfide minerals. However, no single hypothesis completely explains the fate of the As mobilization process in Bangladesh, and most of the hypotheses are valid at the local level. The geology, geomorphology, sediment types, groundwater flow, hydrochemical as well as anthropogenic factors, use of fertilizer, groundwater flow pathway, and adverse effects of irrigation control the occurrence and mobilization of As in Bangladesh. Hence, it is essential to apply every hypothesis to relate to the different areas of Bangladesh. In Bangladesh, a small number of investigations have been conducted on the role of environmental isotopes in the occurrence and mobilization of As. Therefore, a study of the relationship between environmental isotopes and As is required to assess the environmental consequences for human health.

Acknowledgements

This work was supported in part by the National key R & D plan on strategic international scientific and technological innovation cooperation special project under Grant 2016YFE0202300, the National Natural Science Foundation of China under Grants 61671332, 41771452, 51708426 and 41771454, the Natural Science Fund of Hubei Province in China under Grant 2018CFA007, and the Independent Research Projects of Wuhan University under Grant 2042018kf0250.

References

- Abass, O.K., Ma, T., Kong, S., Wang, Z., Mpinda, M.T., 2016. A novel MD-ZVI integrated approach for high arsenic groundwater decontamination and effluent immobilization. *Process Saf. Environ.* 102, 190–203.
- Abedin, M.J., Cresser, M.S., Meharg, A.A., Feldmann, J., Cotter-Howells, J., 2002. Arsenic accumulation and metabolism in rice (*Oryza sativa* L.). *Environ. Sci. Technol.* 36, 962–968.
- Agusa, T., Trang, P.T.K., Lan, V.M., Anh, D.H., Tanabe, S., Viet, P.H., Berg, M., 2014. Human exposure to arsenic from drinking water in Vietnam. *Sci. Total Environ.* 488, 562–569.
- Ahmad, S.A., Sayed, M.H., Khan, M.H., Karim, M.N., Haque, M.A., Bhuiyan, M.S., Rahman, M.S., Faruquee, M.H., 2007. Sociocultural aspects of arsenicosis in Bangladesh: community perspective. *J. Environ. Sci. Heal. A* 42, 1945–1958.
- Ahmed, K.M., Bhattacharya, P., Hasan, M.A., Akhter, S.H., Alam, S.M., Bhuyian, M.H., Imam, M.B., Khan, A.A., Sracek, O., 2004. Arsenic enrichment in groundwater of the alluvial aquifers in Bangladesh: an overview. *Appl. Geochem.* 19, 181–200.
- Ahmed, M.K., Baki, M.A., Kundu, G.K., Islam, M.S., Islam, M.M., Hossain, M.M., 2016a. Human health risks from heavy metals in fish of Buriganga river, Bangladesh. *SpringerPlus* 5, 1697.
- Ahmed, M.K., Shaheen, N., Islam, M.S., Habibullah-Al-Mamun, M., Islam, S., Islam, M.M., Kundu, G.K., Bhattacharjee, L., 2016b. A comprehensive assessment of arsenic in commonly consumed foodstuffs to evaluate the potential health risk in Bangladesh. *Sci. Total Environ.* 544, 125–133.
- Ahmed, S., Akhtar, E., Roy, A., von Ehrenstein, O.S., Vahter, M., Wagatsuma, Y., Raqib, R., 2017. Arsenic exposure alters lung function and airway inflammation in children: a cohort study in rural Bangladesh. *Environ. Int.* 101, 108–116.
- Alam, M., Snow, E., Tanaka, A., 2003. Arsenic and heavy metal contamination of vegetables grown in Samta village, Bangladesh. *Sci. Total Environ.* 308, 83–96.
- Alam, R., McPhedran, K., 2019. Applications of biological sulfate reduction for remediation of arsenic – a review. *Chemosphere* 222, 932–944.
- Alamdar, A., Eqani, S.A.M.A.S., Hanif, N., Ali, S.M., Fasola, M., Bokhari, H., Katsoyiannis, I.A., Shen, H., 2017. Human exposure to trace metals and arsenic via consumption of fish from river Chenab, Pakistan and associated health risks. *Chemosphere* 168, 1004–1012.
- Anawar, H., Akai, J., Mostofa, K., Saifullah, S., Tareq, S., 2002. Arsenic poisoning in groundwater: health risk and geochemical sources in Bangladesh. *Environ. Int.* 27, 597–604.

- Argos, M., Parvez, F., Chen, Y., Hussain, A.I., Momotaj, H., Howe, G.R., Graziano, J.H., Ahsan, H., 2007. Socioeconomic status and risk for arsenic-related skin lesions in Bangladesh. *Am. J. Publ. Health* 97, 825–831.
- Attreed, S.E., Navas-Acien, A., Heaney, C.D., 2017. Arsenic and immune response to infection during pregnancy and early life. *Curr. Environ. Health Rep.* 4, 229–243.
- Ayers, J.C., Goodbred, S., George, G., Fry, D., Benneyworth, L., Hornberger, G., Roy, K., Karim, M.R., Akter, F., 2016. Sources of salinity and arsenic in groundwater in southwest Bangladesh. *Geochem. Trans.* 17, 1–22.
- Azamat, J., Khataee, A., Sadikoglu, F., 2018. Computational study on the efficiency of MoS₂ membrane for removing arsenic from contaminated water. *J. Mol. Liq.* 249, 110–116.
- Aziz, Z., Bostick, B., Zheng, Y., Huq, M., Rahman, M., Ahmed, K., van Geen, A., 2017. Evidence of decoupling between arsenic and phosphate in shallow groundwater of Bangladesh and potential implications. *Appl. Geochem.* 77, 167–177.
- Bahar, M.M., Megharaj, M., Naidu, R., 2016. Oxidation of arsenite to arsenate in growth medium and groundwater using a novel arsenite-oxidizing diazotrophic bacterium isolated from soil. *Int. Biodegrad. Biodegrad.* 106, 178–182.
- Bakhat, H.F., Zia, Z., Fahad, S., Abbas, S., Hammad, H.M., Shahzad, A.N., Abbas, F., Alharbi, H., Shahid, M., 2017. Arsenic uptake, accumulation and toxicity in rice plants: possible remedies for its detoxification: a review. *Environ. Sci. Pollut. Res.* 1–17.
- Banerjee, S., Mahanty, A., Mohanty, S., Mazumder, D.G., Cash, P., Mohanty, B.P., 2017. Identification of potential biomarkers of hepatotoxicity by plasma proteome analysis of arsenic-exposed carp *Labeo rohita*. *J. Hazard Mater.* 336, 71–80.
- Barats, A., Féraud, G., Potot, C., Philippini, V., Travi, Y., Durrieu, G., Dubar, M., Simler, R., 2014. Naturally dissolved arsenic concentrations in the Alpine/Mediterranean Var River watershed (France). *Sci. Total Environ.* 473, 422–436.
- Bassil, M., Daou, F., Hassan, H., Yamani, O., Kharma, J.A., Attieh, Z., Elaridi, J., 2018. Lead, cadmium and arsenic in human milk and their socio-demographic and lifestyle determinants in Lebanon. *Chemosphere* 191, 911–921.
- Bayatkashkoli, A., Kameshki, B., Ravan, S., Shamsian, M., 2017. Comparing of performance of treated particleboard with alkaline copper quat, boron-fluorine-chromium-arsenic and Chlorotalonil against *Microcerotermes diversus* and *Anacanthotermes vagans* termite. *Int. Biodegrad. Biodegrad.* 120, 186–191.
- BBS, UNICEF, 2011. Bangladesh National Drinking Water Quality Survey of 2009. Bangladesh Bureau of Statistics, Dhaka.
- BGS, DPHE, 2001. Arsenic Contamination of Groundwater in Bangladesh. Final Report. BGS Technical Report 2. Bangladesh.
- Bhattacharjee, S., Saha, B., Saha, B., Uddin, M.S., Panna, C.H., Bhattacharya, P., Saha, R., 2019. Groundwater governance in Bangladesh: established practices and recent trends. *Groundwater Sustain. Dev.* 8, 69–81.
- Bhattacharya, P., Hasan, M.A., Sracek, O., Smith, E., Ahmed, K.M., Von Brömssen, M., Huq, S.I., Naidu, R., 2009. Groundwater chemistry and arsenic mobilization in the Holocene flood plains in south-central Bangladesh. *Environ. Geochem. Hlth.* 31, 23–43.
- Bhowmick, S., Pramanik, S., Singh, P., Mondal, P., Chatterjee, D., Nriagu, J., 2018. Arsenic in groundwater of West Bengal, India: a review of human health risks and assessment of possible intervention options. *Sci. Total Environ.* 612, 148–169.
- Bibi, M., Ahmed, F., Ishiga, H., 2008. Geochemical study of arsenic concentrations in groundwater of the Meghna River Delta, Bangladesh. *J. Geochem. Explor.* 97, 43–58.
- Bibi, S., Kamran, M.A., Sultana, J., Farooqi, A., 2017. Occurrence and methods to remove arsenic and fluoride contamination in water. *Environ. Chem. Lett.* 15, 125–149.
- Bodrud-Doza, M., Islam, A.T., Ahmed, F., Das, S., Saha, N., Rahman, M.S., 2016. Characterization of groundwater quality using water evaluation indices, multivariate statistics and geostatistics in central Bangladesh. *Water Sci* 30, 19–40.
- Bortnikova, S., Bessonova, E., Gaskova, O., 2012. Geochemistry of arsenic and metals in stored tailings of a Co-Ni arsenide-ore, Khovu-Aksy area, Russia. *Appl. Geochem.* 27, 2238–2250.
- Bose, R., De, A., 2013. Arsenic contamination: unavoidable natural phenomenon or an anthropogenic crisis. *Proc. Natl. Acad. Sci. India Sect. A (Phys. Sci.): Physical Sciences* 83, 181–185.
- Brima, E.L., Haris, P.I., Jenkins, R.O., Polya, D.A., Gault, A.G., Harrington, C.F., 2006. Understanding arsenic metabolism through a comparative study of arsenic levels in the urine, hair and fingernails of healthy volunteers from three unexposed ethnic groups in the United Kingdom. *Toxicol. Appl. Pharmacol.* 216, 122–130.
- Cao, W., Guo, H., Zhang, Y., Ma, R., Li, Y., Dong, Q., Li, Y., Zhao, R., 2018. Controls of paleochannels on groundwater arsenic distribution in shallow aquifers of alluvial plain in the Hetao Basin, China. *Sci. Total Environ.* 613, 958–968.
- Caporale, A.G., Adamo, P., Azam, S.M., Rao, M.A., Pigna, M., 2018. May humic acids or mineral fertilisation mitigate arsenic mobility and availability to carrot plants (*Daucus carota* L.) in a volcanic soil polluted by as from irrigation water? *Chemosphere* 193, 464–471.
- Chakraborti, D., Rahman, M.M., Das, B., Murrill, M., Dey, S., Mukherjee, S.C., Dhar, R.K., Biswas, B.K., Chowdhury, U.K., Roy, S., 2010. Status of groundwater arsenic contamination in Bangladesh: a 14-year study report. *Water Res.* 44, 5789–5802.
- Chakraborti, D., Rahman, M.M., Mukherjee, A., Alauddin, M., Hassan, M., Dutta, R.N., Pati, S., Mukherjee, S.C., Roy, S., Quamruzzaman, Q., Rahman, M., Morshed, S., Islam, T., Sorif, S., Selim, M., Islam, M.R., Hossain, M.M., 2015. Groundwater arsenic contamination in Bangladesh-21 Years of research. *J. Trace Elem. Med. Biol.* 31, 237–248.
- Chakraborti, D., Rahman, M.M., Paul, K., Chowdhury, U.K., Sengupta, M.K., Lodh, D., Chanda, C.R., Saha, K.C., Mukherjee, S.C., 2002. Arsenic calamity in the Indian subcontinent: what lessons have been learned? *Talanta* 58, 3–22.
- Chakraborty, M., Mukherjee, A., Ahmed, K.M., 2015. A review of groundwater arsenic in the Bengal Basin, Bangladesh and India: from source to sink. *Curr. Pollut. Rep.* 1, 220–247.
- Chan, N., Roy, R., Chaffin, B., 2016. Water governance in Bangladesh: an evaluation of institutional and political context. *Water* 8, 403.
- Chanpiwat, P., Sthiannopkao, S., Cho, K.H., Kim, K.-W., San, V., Suvanthon, B., Vongthavady, C., 2011. Contamination by arsenic and other trace elements of tube-well water along the Mekong River in Lao PDR. *Environ. Pollut.* 159, 567–576.
- Chatterjee, D., Halder, D., Majumder, S., Biswas, A., Nath, B., Bhattacharya, P., Bhowmick, S., Mukherjee-Goswami, A., Saha, D., Hazra, R., 2010. Assessment of arsenic exposure from groundwater and rice in Bengal Delta Region, West Bengal, India. *Water Res.* 44, 5803–5812.
- Chen, X., Zeng, X.-C., Wang, J., Deng, Y., Ma, T., Guo, J., Mu, Y., Yang, Y., Li, H., Wang, Y., 2017. Microbial communities involved in arsenic mobilization and release from the deep sediments into groundwater in Jiangnan plain, Central China. *Sci. Total Environ.* 579, 989–999.
- Cheng, Y.-Y., Huang, N.-C., Chang, Y.-T., Sung, J.-M., Shen, K.-H., Tsai, C.-C., Guo, H.-R., 2017. Associations between arsenic in drinking water and the progression of chronic kidney disease: a nationwide study in Taiwan. *J. Hazard Mater.* 321, 432–439.
- Chowdhury, M., Uddin, M., Ahmed, M., Ali, M., Rasul, S., Hoque, M., Alam, R., Sharmin, R., Uddin, S., Islam, M., 2006. Collapse of Socio-Economic Base of Bangladesh by Arsenic Contamination in Groundwater.
- Chowdhury, M.T.A., Deacon, C.M., Jones, G.D., Imamul Huq, S.M., Williams, P.N., Manzurul Hoque, A.F.M., Winkel, L.H.E., Price, A.H., Norton, G.J., Meharg, A.A., 2017. Arsenic in Bangladeshi soils related to physiographic region, paddy management, and micro- and macro-elemental status. *Sci. Total Environ.* 590–591, 406–415.
- Chowdhury, M.T.A., Deacon, C.M., Steel, E., Imamul Huq, S.M., Paton, G.I., Price, A.H., Williams, P.N., Meharg, A.A., Norton, G.J., 2018. Physiographical variability in arsenic dynamics in Bangladeshi soils. *Sci. Total Environ.* 612, 1365–1372.
- Cui, J.-l., Zhao, Y.-p., Li, J.-s., Beiyuan, J.-z., Tsang, D.C., Poon, C.-s., Chan, T.-s., Wang, W.-x., Li, X.-d., 2018. Speciation, mobilization, and bioaccessibility of arsenic in geogenic soil profile from Hong Kong. *Environ. Pollut.* 232, 375–384.
- Dai, X., Li, P., Tu, J., Zhang, R., Wei, D., Li, B., Wang, Y., Jiang, Z., 2016. Evidence of arsenic mobilization mediated by an indigenous iron reducing bacterium from high arsenic groundwater aquifer in Hetao Basin of Inner Mongolia, China. *Int. Biodegrad. Biodegrad.* <https://doi.org/10.1016/j.ibiod.2016.05.012>.
- Das, H., Mitra, A.K., Sengupta, P., Hossain, A., Islam, F., Rabbani, G., 2004. Arsenic concentrations in rice, vegetables, and fish in Bangladesh: a preliminary study. *Environ. Int.* 30, 383–387.
- Das, N., Das, A., Sarma, K.P., Kumar, M., 2018. Provenance, prevalence and health perspective of co-occurrences of arsenic, fluoride and uranium in the aquifers of the Brahmaputra River floodplain. *Chemosphere* 194, 755–772.
- De, S., Maiti, A., 2011. Arsenic Removal from Contaminated Groundwater Using Laterite Based Adsorption Technique. TERI Press, India.
- Deschamps, E., Ciminelli, V.S., Höll, W.H., 2005. Removal of as (III) and as (V) from water using a natural Fe and Mn enriched sample. *Water Res.* 39, 5212–5220.
- Dhar, R.K., Biswas, B.K., Samanta, G., Mandal, B.K., Chakraborti, D., Roy, S., Jafar, A., Islam, A., Ara, G., Kabir, S., 1997. Groundwater arsenic calamity in Bangladesh. *Curr. Sci.* 48–59.
- Diwakar, J., Johnston, S.G., Burton, E.D., Shrestha, S.D., 2015. Arsenic mobilization in an alluvial aquifer of the Terai region, Nepal. *J. Hydrol. Reg. Stud.* 4, 59–79.
- Donselaar, M.E., Bhatt, A.G., Ghosh, A.K., 2017. On the relation between fluvio-deltaic flood basin geomorphology and the wide-spread occurrence of arsenic pollution in shallow aquifers. *Sci. Total Environ.* 574, 901–913.
- Dousova, B., Buzek, F., Lhotka, M., Krejcová, S., Boubinova, R., 2016. Leaching effect on arsenic mobility in agricultural soils. *J. Hazard Mater.* 307, 231–239.
- Dowling, C.B., Poreda, R.J., Basu, A.R., Peters, S.L., Aggarwal, P.K., 2002. Geochemical study of arsenic release mechanisms in the Bengal Basin groundwater. *Water Resour. Res.* 38.
- Duan, Y., Gan, Y., Wang, Y., Liu, C., Yu, K., Deng, Y., Zhao, K., Dong, C., 2017. Arsenic speciation in aquifer sediment under varying groundwater regime and redox conditions at Jiangnan Plain of Central China. *Sci. Total Environ.* 607, 992–1000.
- Duncan, E.G., Maher, W.A., Foster, S.D., Krikowa, F., O'Sullivan, C.A., Roper, M.M., 2017. Dimethylarsenate (DMA) exposure influences germination rates, arsenic uptake and arsenic species formation in wheat. *Chemosphere* 181, 44–54.
- Edmunds, W., Ahmed, K., Whitehead, P., 2015. A review of arsenic and its impacts in groundwater of the Ganges-Brahmaputra-Meghna delta, Bangladesh. *Environ. Sci. Proc. Imp.* 17, 1032–1046.
- Fang, T.-H., Chen, Y.-S., 2015. Arsenic speciation and diffusion flux in danshuei estuary sediments, northern Taiwan. *Mar. Pollut. Bull.* 101, 98–109.
- Fendorf, S., Michael, H.A., van Geen, A., 2010. Spatial and temporal variations of groundwater arsenic in South and Southeast Asia. *Science* 328, 1123–1127.
- Flora, S.J.S., 2015. 1 - arsenic: chemistry, occurrence, and exposure. In: Flora, S.J.S. (Ed.), *Handbook of Arsenic Toxicology*. Academic Press, Oxford, pp. 1–49.
- Fransisca, Y., Small, D.M., Morrison, P.D., Spencer, M.J., Ball, A.S., Jones, O.A., 2015. Assessment of arsenic in Australian grown and imported rice varieties on sale in Australia and potential links with irrigation practices and soil geochemistry. *Chemosphere* 138, 1008–1013.
- Freikowsky, D., Neidhardt, H., Winter, J., Berner, Z., Gallert, C., 2013. Effect of carbon sources and of sulfate on microbial arsenic mobilization in sediments of West Bengal, India. *Ecotoxicol. Environ. Saf.* 91, 139–146.
- Galbacs, M., Galbacs, G., 1995. International Conference on Arsenic in Ground Water: Cause, Effect and Remedy (Feb).
- Gao, X., Su, C., Wang, Y., Hu, Q., 2013. Mobility of arsenic in aquifer sediments at Datong Basin, northern China: effect of bicarbonate and phosphate. *J. Geochem. Explor.* 135, 93–103.

- Garshick, M., Wu, F., Demmer, R., Parvez, F., Ahmed, A., Eunus, M., Hasan, R., Nahar, J., Shaheen, I., Sarwar, G., 2017. The association between socioeconomic status and subclinical atherosclerosis in a rural Bangladesh population. *Prev. Med.* 102, 6–11.
- Goel, V., Islam, M., Yunus, M., Ali, M., Khan, A., Alam, N., Faruque, A., Bell, G., Sobsey, M., Emch, M., 2019. Deep tubewell microbial water quality and access in arsenic mitigation programs in rural Bangladesh. *Sci. Total Environ.* 659, 1577–1584.
- Gomez-Gonzalez, M.A., Voegelin, A., Garcia-Guinea, J., Bolea, E., Laborda, F., Garrido, F., 2016. Colloidal mobilization of arsenic from mining-affected soils by surface runoff. *Chemosphere* 144, 1123–1131.
- Gong, X.-J., Li, W.-G., Zhang, D.-Y., Fan, W.-B., Zhang, X.-R., 2015. Adsorption of arsenic from micro-polluted water by an innovative coal-based mesoporous activated carbon in the presence of co-existing ions. *Int. Biodeterior. Biodegrad.* 102, 256–264.
- Guo, H.-R., Tseng, Y.-C., 2000. Arsenic in drinking water and bladder cancer: comparison between studies based on cancer registry and death certificates. *Environ. Geochem. Hlth.* 22, 83–91.
- Guo, H., Liu, Z., Ding, S., Hao, C., Xiu, W., Hou, W., 2015. Arsenate reduction and mobilization in the presence of indigenous aerobic bacteria obtained from high arsenic aquifers of the Hetao basin, Inner Mongolia. *Environ. Pollut.* 203, 50–59.
- Guo, H., Stüben, D., Berner, Z., 2007. Arsenic removal from water using natural iron mineral-quartz sand columns. *Sci. Total Environ.* 377, 142–151.
- Guo, H., Zhang, D., Ni, P., Cao, Y., Li, F., 2017. Hydrogeological and geochemical comparison of high arsenic groundwaters in inland basins, PR China. *Proced. Earth Plan. Sci.* 17, 416–419.
- Guo, Q., Guo, H., Yang, Y., Han, S., Zhang, F., 2014. Hydrogeochemical contrasts between low and high arsenic groundwater and its implications for arsenic mobilization in shallow aquifers of the northern Yinchuan Basin, PR China. *J. Hydrol.* 518, 464–476.
- Gupta, D.K., Srivastava, S., Huang, H., Romero-Puertas, M.C., Sandalo, L.M., 2011. Arsenic Tolerance and Detoxification Mechanisms in Plants, Detoxification of Heavy Metals. Springer, pp. 169–179.
- Hadi, A., Parveen, R., 2004. Arsenicosis in Bangladesh: prevalence and socio-economic correlates. *Publ. Health* 118, 559–564.
- Haque, M.N., Keramat, M., Shahid, S., 2015. Groundwater resource evaluation in the western part of Kushtia district of Bangladesh using vertical electrical sounding technique. *ISH J. Hydraul. Eng.* 21, 97–110.
- Haque, M.N., Keramat, M., Shahid, S., Mohsenipour, M., Wang, X.-J., 2017. Groundwater dynamics and balance in the western part of greater Kushtia district of Bangladesh. *KSCE J. Civil Engin* 21, 1595–1606.
- Hare, V., Chowdhury, P., Baghel, V.S., 2017. Influence of bacterial strains on *Oryza sativa* grown under arsenic tainted soil: accumulation and detoxification response. *Plant Physiol. Biochem. (Paris)* 119, 93–102.
- Harvey, C.F., Ashfaq, K.N., Yu, W., Badruzzaman, A., Ali, M.A., Oates, P.M., Michael, H.A., Neumann, R.B., Beckie, R., Islam, S., 2006. Groundwater dynamics and arsenic contamination in Bangladesh. *Chem. Geol.* 228, 112–136.
- Hasan, M.A., Bhattacharya, P., Sracek, O., Ahmed, K.M., von Brömssen, M., Jacks, G., 2009. Geological controls on groundwater chemistry and arsenic mobilization: hydrogeochemical study along an E-W transect in the Meghna basin, Bangladesh. *J. Hydrol.* 378, 105–118.
- Hassan, M.M., Atkins, P.J., Dunn, C.E., 2003. The spatial pattern of risk from arsenic poisoning: a Bangladesh case study. *J. Environ. Sci. Heal. A* 38, 1–24.
- Hassan, M.M., Atkins, P.J., Dunn, C.E., 2005. Social implications of arsenic poisoning in Bangladesh. *Soc. Sci. Med.* 61, 2201–2211.
- Herath, I., Vithanage, M., Bundschuh, J., Maity, J.P., Bhattacharya, P., 2016. Natural arsenic in global groundwaters: distribution and geochemical triggers for mobilization. *Curr. Pollut. Rep.* 2, 68–89.
- Hossain, M., 2006. Arsenic contamination in Bangladesh—an overview. *Agric. Ecosyst. Environ.* 113, 1–16.
- Hossain, M.G., Reza, A.S., Lutfun-Nessa, M., Ahmed, S.S., 2013. Factor and cluster analysis of water quality data of the groundwater wells of Kushtia, Bangladesh: implication for arsenic enrichment and mobilization. *J. Geol. Soc. India* 81, 377–384.
- Howladar, M.F., 2017. An assessment of surface water chemistry with its possible sources of pollution around the Barapukuria Thermal Power Plant Impacted Area, Dinajpur, Bangladesh. *Groundwater Sustain. Dev.* 5, 38–48.
- Hsueh, Y.-M., Su, C.-T., Shiue, H.-S., Chen, W.-J., Pu, Y.-S., Lin, Y.-C., Tsai, C.-S., Huang, C.-Y., 2017. Levels of plasma selenium and urinary total arsenic interact to affect the risk for prostate cancer. *Food Chem. Toxicol.* 107, 167–175.
- Huq, M.E., Fahad, S., Shao, Z., Sarven, M.S., Al-Huqail, A.A., Siddiqui, M.H., Rahman, M. H.u., Khan, I.A., Alam, M., Saeed, M., Rauf, A., Basir, A., Jamal, Y., Khan, S.U., 2019. High arsenic contamination and presence of other trace metals in drinking water of Kushtia district, Bangladesh. *J. Environ. Manag.* 242, 199–209.
- Huq, M.E., Su, C., Fahad, S., Li, J., Sarven, M.S., Liu, R., 2018. Distribution and hydrogeochemical behavior of arsenic enriched groundwater in the sedimentary aquifer comparison between Datong Basin (China) and Kushtia District (Bangladesh). *Environ. Sci. Pollut. Res.* 25 (16), 15830–15843.
- Huq, M.E., Su, C., Li, J., Sarven, M.S., 2018b. Arsenic enrichment and mobilization in the Holocene alluvial aquifers of Prayagpur of Southwestern Bangladesh. *Int. Biodeterior. Biodegrad.* 128, 186–194.
- Huq, S.I., Rahman, A., Sultana, N., Naidu, R., 2003. Extent and Severity of Arsenic Contamination in Soils of Bangladesh. *Fate of Arsenic in the Environment*. Bangladesh University of Engineering and Technology, Dhaka, pp. 69–84.
- Hussain, M., Abdullah, S., 2001. Geological Setting of the Areas of Arsenic Safe Aquifers. Report of the Ground Water Task Force (Interim report).
- Islam, A.R.M.T., Ahmed, N., Bodrud-Doza, M., Chu, R., 2017a. Characterizing groundwater quality ranks for drinking purposes in Sylhet district, Bangladesh, using entropy method, spatial autocorrelation index, and geostatistics. *Environ. Sci. Pollut. Res.* 1–25.
- Islam, A.T., Sein, Z., Ongoma, V., Islam, M.S., Alam, M.F., Ahmed, F., 2015. Geomorphological and land use mapping: a case study of ishwardi under Pabna district, Bangladesh. *Adv. Res.* 4, 378–387.
- Islam, A.T., Shen, S.-H., Bodrud-Doza, M., 2017b. Assessment of arsenic health risk and source apportionment of groundwater pollutants using multivariate statistical techniques in Chapai-Nawabganj district, Bangladesh. *J. Geol. Soc. India* 90, 239–248.
- Islam, A.T., Shen, S., Bodrud-Doza, M., Rahman, M.A., Das, S., 2017c. Assessment of trace elements of groundwater and their spatial distribution in Rangpur district, Bangladesh. *Arab. J. Geosci.* 10, 95.
- Islam, F.S., Gault, A.G., Boothman, C., Polya, D.A., Charnock, J.M., Chatterjee, D., Lloyd, J.R., 2004. Role of metal-reducing bacteria in arsenic release from Bengal delta sediments. *Nature* 430, 68.
- Islam, M.A., Rahman, M.M., Bodrud-Doza, M., Muhib, M.I., Shammi, M., Zahid, A., Akter, Y., Kurasaki, M., 2017d. A study of groundwater irrigation water quality in south-central Bangladesh: a geo-statistical model approach using GIS and multivariate statistics. *Acta Geochimica* 1–22.
- Islam, M.S., Ahmed, M.K., Habibullah-Al-Mamun, M., Eaton, D.W., 2017e. Arsenic in the food chain and assessment of population health risks in Bangladesh. *Environ. Sys. Dec.* 1–9.
- Islam, S.D.-U., Majumder, R.K., Uddin, M.J., Khalil, M.I., Alam, M.F., 2017f. Hydrochemical characteristics and quality assessment of groundwater in patuakhali district, southern coastal region of Bangladesh. *Expos. Health* 9, 43–60.
- Joseph, T., Dubey, B., McBean, E.A., 2015a. A critical review of arsenic exposures for Bangladeshi adults. *Sci. Total Environ.* 527, 540–551.
- Joseph, T., Dubey, B., McBean, E.A., 2015b. Human health risk assessment from arsenic exposures in Bangladesh. *Sci. Total Environ.* 527, 552–560.
- Jung, H.B., Zheng, Y., Rahman, M.W., Rahman, M.M., Ahmed, K.M., 2015. Redox zonation and oscillation in the hyporheic zone of the Ganges-Brahmaputra-Meghna Delta: implications for the fate of groundwater arsenic during discharge. *Appl. Geochem.* 63, 647–660.
- Kabir, F., Chowdhury, S., 2017. Arsenic removal methods for drinking water in the developing countries: technological developments and research needs. *Environ. Sci. Pollut. Res.* 24, 24102–24120.
- Kanoua, W., Merkel, B., 2017. Hydrochemical evolution and arsenic release in shallow aquifer in the Titas Upazila, Eastern Bangladesh. *Arab. J. Geosci.* 10, 290.
- Khan, N.I., Owens, G., Bruce, D., Naidu, R., 2009. Human arsenic exposure and risk assessment at the landscape level: a review. *Environ. Geochem. Hlth.* 31, 143.
- Kile, M.L., Faraj, J.M., Ronnenberg, A.G., Quamruzzaman, Q., Rahman, M., Mostofa, G., Afroz, S., Christiani, D.C., 2016. A cross sectional study of anemia and iron deficiency as risk factors for arsenic-induced skin lesions in Bangladeshi women. *BMC Publ. Health* 16, 158.
- Kile, M.L., Rodrigues, E.G., Mazumdar, M., Dobson, C.B., Diao, N., Golam, M., Quamruzzaman, Q., Rahman, M., Christiani, D.C., 2014. A prospective cohort study of the association between drinking water arsenic exposure and self-reported maternal health symptoms during pregnancy in Bangladesh. *Environ. Health* 13, 29.
- Kinniburgh, D., Smedley, P., 2001. Final Report. Arsenic Contamination of Groundwater in Bangladesh, vol. 2. British Geological Survey. BGS Technical Report No WC/00/19.
- Kinniburgh, D.G., Smedley, P.L., Davies, J., Milne, C.J., Gaus, I., Trafford, J.M., Burden, S., Ihtishamul Huq, S.M., Ahmad, N., Ahmed, K.M., 2003. The scale and causes of the groundwater arsenic problem in Bangladesh. In: Welch, A.H., Stollenwerk, K.G. (Eds.), *Arsenic in Ground Water: Geochemistry and Occurrence*. Springer US, Boston, MA, pp. 211–257.
- Kippler, M., Skroder, H., Rahman, S.M., Tofail, F., Vahter, M., 2016. Elevated childhood exposure to arsenic despite reduced drinking water concentrations—a longitudinal cohort study in rural Bangladesh. *Environ. Int.* 86, 119–125.
- Knappett, P., Mailloux, B., Choudhury, I., Khan, M., Michael, H., Barua, S., Mondal, D., Steckler, M., Akhter, S., Ahmed, K., 2016. Vulnerability of low-arsenic aquifers to municipal pumping in Bangladesh. *J. Hydrol.* 539, 674–686.
- Kulkarni, H.V., Mladenov, N., McKnight, D.M., Zheng, Y., Kirk, M.F., Nemerugut, D.R., 2018. Dissolved fulvic acids from a high arsenic aquifer shuttle electrons to enhance microbial iron reduction. *Sci. Total Environ.* 615, 1390–1395.
- Kumar, M., Rahman, M.M., Ramanathan, A., Naidu, R., 2016. Arsenic and other elements in drinking water and dietary components from the middle Gangetic plain of Bihar, India: health risk index. *Sci. Total Environ.* 539, 125–134.
- Kurzus-Spencer, M., da Silva, V., Thomson, C.A., Hartz, V., Hsu, C.-H., Burgess, J.L., O'Rourke, M.K., Harris, R.B., 2017. Nutrients in one-carbon metabolism and urinary arsenic methylation in the national health and nutrition examination survey (NHANES) 2003–2004. *Sci. Total Environ.* 607, 381–390.
- Lee, J.J., Kapur, K., Rodrigues, E.G., Hasan, M.O.S.I., Quamruzzaman, Q., Wright, R.O., Belling, D.C., Christiani, D.C., Mazumdar, M., 2017. Anthropometric measures at birth and early childhood are associated with neurodevelopmental outcomes among Bangladeshi children aged 2–3 years. *Sci. Total Environ.* 607, 475–482.
- Li, J., Wang, Y., Xie, X., 2016. Cl/Br ratios and chlorine isotope evidences for groundwater salinization and its impact on groundwater arsenic, fluoride and iodine enrichment in the Datong basin. *China. Sci. Total Environ.* 544, 158–167.
- Lin, T.Y., Hafeezezami, S., Rice, L., Lee, J., Maki, A., Sevilla, T., Stahl, M., Neumann, R., Harvey, C., Suflet, I.M., 2017. Arsenic oxyanion binding to NOM from dung and aquaculture pond sediments in Bangladesh: importance of site-specific binding constants. *Appl. Geochem.* 78, 234–240.
- Liu, F., Zhang, G., Liu, S., Fu, Z., Chen, J., Ma, C., 2018. Bioremoval of arsenic and antimony from wastewater by a mixed culture of sulfate-reducing bacteria using lactate and ethanol as carbon sources. *Int. Biodeterior. Biodegrad.* 126, 152–159.

- Liu, J., Gao, Y., Liu, H., Sun, J., Liu, Y., Wu, J., Li, D., Sun, D., 2017. Assessment of relationship on excess arsenic intake from drinking water and cognitive impairment in adults and elders in arsenicosis areas. *Int. J. Hyg Environ. Health* 220, 424–430.
- Liu, Q., Guo, H., Li, Y., Xiang, H., 2013. Acclimation of arsenic-resistant Fe(II)-oxidizing bacteria in aqueous environment. *Int. Biodeterior. Biodegrad.* 76, 86–91.
- Lowers, H.A., Breit, G.N., Foster, A.L., Whitney, J., Yount, J., Uddin, M.N., Muneem, A. A., 2007. Arsenic incorporation into authigenic pyrite, Bengal Basin sediment, Bangladesh. *Geochem. Cosmochim. Acta* 71, 2699–2717.
- Luong, V.T., Cañas Kurz, E.E., Hellriegel, U., Luu, T.L., Hoinkis, J., Bundschuh, J., 2018. Iron-based subsurface arsenic removal technologies by aeration: a review of the current state and future prospects. *Water Res.* 133, 110–122.
- Mahmood, S.A.I., Halder, A.K., 2011. The socioeconomic impact of Arsenic poisoning in Bangladesh. *J. Toxicol. Environ. Health Sci.* 3, 65–73.
- Mahmud, I., Mbuya, N., 2015. Water, Sanitation, Hygiene, and Nutrition in Bangladesh: Can Building Toilets Affect Children's Growth? The World Bank.
- Maier, M.V., Isenbeck-Schröter, M., Klose, L.B., Ritter, S.M., Scholz, C., 2017. In situ mobilization of arsenic in groundwater—an innovative remediation approach? *Proced. Earth Plan. Sc.* 17, 452–455.
- Maizel, D., Blum, J.S., Ferrero, M.A., Utturkar, S.M., Brown, S.D., Rosen, B.P., Oremland, R.S., 2016. Characterization of the extremely arsenic-resistant *Brevibacterium linens* strain AE038-8 isolated from contaminated groundwater in Tucumán, Argentina. *Int. Biodeterior. Biodegrad.* 107, 147–153.
- Majumder, R., Halim, M., Shimada, J., Saha, B., Zahid, A., Hasan, M., Islam, M., 2013. Hydrochemistry and isotopic studies to identify Ganges River and riverbank groundwater interaction, southern Bangladesh. *Arab. J. Geosci.* 6, 4585–4591.
- Mandal, A., Purakayastha, T., Patra, A., Sarkar, B., 2017. Arsenic phytoextraction by *Pteris vittata* improves microbial properties in contaminated soil under various phosphate fertilizations. *Appl. Geochem.* <https://doi.org/10.1016/j.apgeochem.2017.04.008>.
- Mandal, B.K., Suzuki, K.T., 2002. Arsenic round the world: a review. *Talanta* 58, 201–235.
- Mango, H., Ryan, P., 2015. Source of arsenic-bearing pyrite in southwestern Vermont, USA: sulfur isotope evidence. *Sci. Total Environ.* 505, 1331–1339.
- Martin, M., Stanchi, S., Hossain, K.J., Huq, S.I., Barberis, E., 2015. Potential phosphorus and arsenic mobilization from Bangladesh soils by particle dispersion. *Sci. Total Environ.* 536, 973–980.
- Martínez-Villegas, N., Briones-Gallardo, R., Ramos-Leal, J.A., Avalos-Borja, M., Castañón-Sandoval, A.D., Razo-Flores, E., Villalobos, M., 2013. Arsenic mobility controlled by solid calcium arsenates: a case study in Mexico showcasing a potentially widespread environmental problem. *Environ. Pollut.* 176, 114–122.
- Mass, M.J., Tennant, A., Roop, B.C., Cullen, W.R., Styblo, M., Thomas, D.J., Kligerman, A.D., 2001. Methylated trivalent arsenic species are genotoxic. *Chem. Res. Toxicol.* 14, 355–361.
- Mateen, F.J., Grau-Perez, M., Pollak, J.S., Moon, K.A., Howard, B.V., Umans, J.G., Best, L.G., Francesconi, K.A., Goessler, W., Crainiceanu, C., 2017. Chronic arsenic exposure and risk of carotid artery disease: the Strong Heart Study. *Environ. Res.* 157, 127–134.
- Mazumdar, M., Christiani, D.C., Golam, M., Quamruzzaman, Q., Dobson, C.B., Rahman, M., Diao, N., Kile, M.L., Rodrigues, E.G., 2014. A Prospective Cohort Study of the Association between Drinking Water Arsenic Exposure and Self-Reported Maternal Health Symptoms during Pregnancy in Bangladesh.
- McArthur, J., Ravenscroft, P., Safiulla, S., Thirlwall, M., 2001. Arsenic in groundwater: testing pollution mechanisms for sedimentary aquifers in Bangladesh. *Water Resour. Res.* 37, 109–117.
- Meng, X., Dupont, R.R., Sorensen, D.L., Jacobson, A.R., McLean, J.E., 2017. Mineralogy and geochemistry affecting arsenic solubility in sediment profiles from the shallow basin-fill aquifer of Cache Valley Basin, Utah. *Appl. Geochem.* 77, 126–141.
- Michael, H.A., Khan, M.R., 2016. Impacts of physical and chemical aquifer heterogeneity on basin-scale solute transport: vulnerability of deep groundwater to arsenic contamination in Bangladesh. *Adv. Water Resour.* 98, 147–158.
- Milosković, A., Simić, V., 2015. Arsenic and other trace elements in five edible fish species in relation to fish size and weight and potential health risks for human consumption. *Pol. J. Environ. Stud.* 24, 199–206.
- Minatel, B.C., Sage, A.P., Anderson, C., Hubaux, R., Marshall, E.A., Lam, W.L., Martinez, V.D., 2018. Environmental arsenic exposure: from genetic susceptibility to pathogenesis. *Environ. Int.* 112, 183–197.
- Mondal, S., Roy, A., Mukherjee, R., Mondal, M., Karmakar, S., Chatterjee, S., Mukherjee, M., Bhattacharjee, S., De, S., 2017. A socio-economic study along with impact assessment for laterite based technology demonstration for arsenic mitigation. *Sci. Total Environ.* 583, 142–152.
- Mosler, H.-J., Blöchliger, O.R., Inauen, J., 2010. Personal, social, and situational factors influencing the consumption of drinking water from arsenic-safe deep tubewells in Bangladesh. *J. Environ. Manag.* 91, 1316–1323.
- Mukherjee, A., Fryar, A.E., 2008. Deeper groundwater chemistry and geochemical modeling of the arsenic affected western Bengal basin, West Bengal, India. *Appl. Geochem.* 23, 863–894.
- Mukherjee, A., Fryar, A.E., Scanlon, B.R., Bhattacharya, P., Bhattacharya, A., 2011. Elevated arsenic in deeper groundwater of the western Bengal basin, India: extent and controls from regional to local scale. *Appl. Geochem.* 26, 600–613.
- Mukherjee, A., Kundu, M., Basu, B., Sinha, B., Chatterjee, M., Bairagya, M.D., Singh, U., Sarkar, S., 2017. Arsenic load in rice ecosystem and its mitigation through deficit irrigation. *J. Environ. Manag.* 197, 89–95.
- Mukherjee, A., Verma, S., Gupta, S., Henke, K.R., Bhattacharya, P., 2014. Influence of tectonics, sedimentation and aqueous flow cycles on the origin of global groundwater arsenic: paradigms from three continents. *J. Hydrol.* 518, 284–299.
- Mukherjee, P., Chatterjee, D., Jana, J., Maity, P.B., Goswami, A., Saha, H., Sen, M., Nath, B., Shome, D., Joyti Sarkar, M., Bagchi, D., 2007. Household Water Treatment Option: Removal of Arsenic in Presence of Natural Fe-Containing Groundwater by Solar Oxidation, Trace Metals and Other Contaminants in the Environment. Elsevier, pp. 603–622.
- Mukhopadhyay, R., Manjiaah, K., Datta, S., Yadav, R., Sarkar, B., 2017. Inorganically modified clay minerals: preparation, characterization, and arsenic adsorption in contaminated water and soil. *Appl. Clay Sci.* 147, 1–10.
- Munoz, M.O., Wern, H., Johnsson, F., Bhattacharya, P., Sracek, O., Thunvik, R., Quintanilla, J., Bundschuh, J., 2013. Geogenic arsenic and other trace elements in the shallow hydrogeologic system of Southern Poopó Basin, Bolivian Altiplano. *J. Hazard Mater.* 262, 924–940.
- Nahar, N., 2009. Impacts of arsenic contamination in groundwater: case study of some villages in Bangladesh. *Environ. Dev. Sustain.* 11, 571–588.
- Nath, B., Sahu, S.J., Jana, J., Mukherjee-Goswami, A., Roy, S., Sarkar, M.J., Chatterjee, D., 2008. Hydrochemistry of arsenic-enriched aquifer from rural West Bengal, India: a study of the arsenic exposure and mitigation option. *Water Air Soil Pollut.* 190, 95–113.
- Nazari, A.M., Radzinski, R., Ghahreman, A., 2017. Review of arsenic metallurgy: treatment of arsenical minerals and the immobilization of arsenic. *Hydrometallurgy* 174, 258–281.
- Neidhardt, H., Winkel, L.H., Kaegi, R., Stengel, C., Trang, P.T., Lan, V.M., Viet, P.H., Berg, M., 2018. Insights into arsenic retention dynamics of Pleistocene aquifer sediments by in situ sorption experiments. *Water Res.* 129, 123–132.
- Ng, J.C., Wang, J., Shraim, A., 2003. A global health problem caused by arsenic from natural sources. *Chemosphere* 52, 1353–1359.
- Ohno, K., Yanase, T., Matsuo, Y., Kimura, T., Rahman, M.H., Magara, Y., Matsui, Y., 2007. Arsenic intake via water and food by a population living in an arsenic-affected area of Bangladesh. *Sci. Total Environ.* 381, 68–76.
- Ourshalimian, S., Naser, A.M., Rahman, M., Doza, S., Stowell, J., Narayan, K.M.V., Shamsudduha, M., Gribble, M.O., 2019. Arsenic and fasting blood glucose in the context of other drinking water chemicals: a cross-sectional study in Bangladesh. *Environ. Res.* 172, 249–257.
- Parvez, F., Medina, S., Santella, R.M., Islam, T., Lauer, F.T., Alam, N., Eunus, M., Rahman, M., Factor-Litvak, P., Ahsan, H., Graziano, J.H., Liu, K.J., Burchiel, S.W., 2017. Arsenic exposures alter clinical indicators of anemia in a male population of smokers and non-smokers in Bangladesh. *Toxicol. Appl. Pharmacol.* 331, 62–68.
- Paul, B.K., 2004. Arsenic contamination awareness among the rural residents in Bangladesh. *Soc. Sci. Med.* 59, 1741–1755.
- Pell, A., Kokkinis, G., Malea, P., Pergantis, S.A., Rubio, R., López-Sánchez, J.F., 2013. LC-ICP-MS analysis of arsenic compounds in dominant seaweeds from the Theraikos Gulf (Northern Aegean Sea, Greece). *Chemosphere* 93, 2187–2194.
- Petrick, J.S., Ayala-Fierro, F., Cullen, W.R., Carter, D.E., Aposhian, H.V., 2000. Monomethylarsonous acid (MMAII) is more toxic than arsenite in Chang human hepatocytes. *Toxicol. Appl. Pharmacol.* 163, 203–207.
- Pi, K., Wang, Y., Xie, X., Liu, Y., Ma, T., Su, C., 2016. Multilevel hydrogeochemical monitoring of spatial distribution of arsenic: a case study at Datong Basin, northern China. *J. Geochem. Explor.* 161, 16–26.
- Pi, K., Wang, Y., Xie, X., Ma, T., Liu, Y., Su, C., Zhu, Y., Wang, Z., 2017a. Remediation of arsenic-contaminated groundwater by in-situ stimulating biogenic precipitation of iron sulfides. *Water Res.* 109, 337–346.
- Pi, K., Wang, Y., Xie, X., Ma, T., Su, C., Liu, Y., 2017b. Role of sulfur redox cycling on arsenic mobilization in aquifers of Datong Basin, northern China. *Appl. Geochem.* 77, 31–43.
- Pi, K., Wang, Y., Xie, X., Su, C., Ma, T., Li, J., Liu, Y., 2015. Hydrogeochemistry of co-occurring geogenic arsenic, fluoride and iodine in groundwater at Datong Basin, northern China. *J. Hazard Mater.* 300, 652–661.
- Polizzotto, M.L., Harvey, C.F., Li, G., Badruzzaman, B., Ali, A., Newville, M., Sutton, S., Fendorf, S., 2006. Solid-phases and desorption processes of arsenic within Bangladesh sediments. *Chem. Geol.* 228, 97–111.
- Polizzotto, M.L., Lineberger, E.M., Matteson, A.R., Neumann, R.B., Badruzzaman, A.B.M., Ali, M.A., 2013. Arsenic transport in irrigation water across rice-field soils in Bangladesh. *Environ. Pollut.* 179, 210–217.
- Postma, D., Trang, P.T.K., Sø, H.U., Lan, V.M., Jakobsen, R., 2017. Reactive transport modeling of arsenic mobilization in groundwater of the red river floodplain, vietnam. *Proced. Earth Plan. Sc.* 17, 85–87.
- Radloff, K.A., Cheng, Z., Rahman, M.W., Ahmed, K.M., Mailloux, B.J., Juhl, A.R., Schlosser, P., van Geen, A., 2007. Mobilization of arsenic during one-year incubations of grey aquifer sands from Araihaaz, Bangladesh. *Environ. Sci. Technol.* 41, 3639–3645.
- Radloff, K.A., Zheng, Y., Stute, M., Weinman, B., Bostick, B., Mihajlov, I., Bounds, M., Rahman, M.M., Huq, M.R., Ahmed, K.M., Schlosser, P., van Geen, A., 2017. Reversible adsorption and flushing of arsenic in a shallow, Holocene aquifer of Bangladesh. *Appl. Geochem.* 77, 142–157.
- Rahman, A., Vahter, M., Ekström, E.C., Persson, L.A., 2011. Arsenic exposure in pregnancy increases the risk of lower respiratory tract infection and diarrhea during infancy in Bangladesh. *Environ. Health Perspect.* 119, 719.
- Rahman, M.A., Rahman, A., Khan, M.Z.K., Renzaho, A.M.N., 2018a. Human health risks and socio-economic perspectives of arsenic exposure in Bangladesh: a scoping review. *Ecotoxicol. Environ. Saf.* 150, 335–343.
- Rahman, M.L., Kile, M.L., Rodrigues, E.G., Valeri, L., Raj, A., Mazumdar, M., Mostofa, G., Quamruzzaman, Q., Rahman, M., Hauser, R., 2018b. Prenatal arsenic exposure, child marriage, and pregnancy weight gain: associations with preterm birth in Bangladesh. *Environ. Int.* 112, 23–32.

- Rahman, M.M., Dong, Z., Naidu, R., 2015. Concentrations of arsenic and other elements in groundwater of Bangladesh and West Bengal, India: potential cancer risk. *Chemosphere* 139, 54–64.
- Rahman, M.M., Sultana, R., Shhami, M., Bikash, J., Ahmed, T., Maruo, M., Kurasaki, M., Uddin, M.K., 2016a. Assessment of the status of groundwater arsenic at Singair upazila, Manikganj Bangladesh; exploring the correlation with other metals and ions. *Expos. Health* 8, 217–225.
- Rahman, M.T.U., Siddiqi, U.R., Kure, S., Mano, A., Udo, K., Ishibashi, Y., 2016b. Mobilization of high arsenic in the shallow groundwater of Kalaroa, south-western Bangladesh. *Expos. Health* 8, 159–175.
- Ramos, O.E.R., Rötting, T.S., French, M., Sracek, O., Bundschuh, J., Quintanilla, J., Bhattacharya, P., 2014. Geochemical processes controlling mobilization of arsenic and trace elements in shallow aquifers and surface waters in the Antequera and Poopó mining regions, Bolivian Altiplano. *J. Hydrol* 518, 421–433.
- Rasul, S.B., Munir, A., Hossain, Z., Khan, A., Alauddin, M., Hussam, A., 2002. Electrochemical measurement and speciation of inorganic arsenic in groundwater of Bangladesh. *Talanta* 58, 33–43.
- Reyes, F.A.P., Crosta, G.B., Frattini, P., Basiricò, S., Della Pergola, R., 2015. Hydrogeochemical overview and natural arsenic occurrence in groundwater from alpine springs (upper Valtellina, Northern Italy). *J. Hydrol* 529, 1530–1549.
- Reza, A.S., Jean, J.-S., 2012. Vertical distribution and mobilization of arsenic in shallow alluvial aquifers of Chapai-Nawabganj district, Northwestern Bangladesh. *J. Geol. Soc. India* 80, 531–538.
- Reza, A.S., Jean, J.-S., Lee, M.-K., Liu, C.-C., Bundschuh, J., Yang, H.-J., Lee, J.-F., Lee, Y.-C., 2010a. Implications of organic matter on arsenic mobilization into groundwater: evidence from northwestern (Chapai-Nawabganj), central (Manikganj) and southeastern (Chandpur) Bangladesh. *Water Res.* 44, 5556–5574.
- Reza, A.S., Jean, J.-S., Yang, H.-J., Lee, M.-K., Woodall, B., Liu, C.-C., Lee, J.-F., Luo, S.-D., 2010b. Occurrence of arsenic in core sediments and groundwater in the Chapai-Nawabganj District, northwestern Bangladesh. *Water Res.* 44, 2021–2037.
- Roberts, L.C., Hug, S.J., Dittmar, J., Voegelin, A., Kretzschmar, R., Wehrli, B., Cirpka, O. A., Saha, G.C., Ali, M.A., Badruzzaman, A.B.M., 2010. Arsenic release from paddy soils during monsoon flooding. *Nat. Geosci.* 3, 53.
- Saha, J., Dikshit, A., Bandyopadhyay, M., Saha, K., 1999. A review of arsenic poisoning and its effects on human health. *Crit. Rev. Environ. Sci. Technol.* 29, 281–313.
- Saint-Jacques, N., Brown, P., Nauta, L., Boxall, J., Parker, L., Dummer, T.J., 2018. Estimating the risk of bladder and kidney cancer from exposure to low-levels of arsenic in drinking water, Nova Scotia, Canada. *Environ. Int.* 110, 95–104.
- Schaefer, M.V., Guo, X., Gan, Y., Benner, S.G., Griffin, A.M., Gorski, C.A., Wang, Y., Fendorf, S., 2017. Redox controls on arsenic enrichment and release from aquifer sediments in central Yangtze River Basin. *Geochem. Cosmochim. Acta* 204, 104–119.
- Shamsudduha, M., Uddin, A., Saunders, J., Lee, M.-K., 2008. Quaternary stratigraphy, sediment characteristics and geochemistry of arsenic-contaminated alluvial aquifers in the Ganges–Brahmaputra floodplain in central Bangladesh. *J. Contam. Hydrol.* 99, 112–136.
- Sharma, A., Flora, S.J.S., 2018. Nutritional management can assist a significant role in alleviation of arsenicosis. *J. Trace Elem. Med. Biol.* 45, 11–20.
- Sharma, A.K., Tjell, J.C., Sloth, J.J., Holm, P.E., 2014. Review of arsenic contamination, exposure through water and food and low cost mitigation options for rural areas. *Appl. Geochem.* 41, 11–33.
- Sharma, V.K., Sohn, M., 2009. Aquatic arsenic: toxicity, speciation, transformations, and remediation. *Environ. Int.* 35, 743–759.
- Shih, Y.-H., Islam, T., Hore, S.K., Sarwar, G., Shahriar, M.H., Yunus, M., Graziano, J.H., Harjes, J., Baron, J.A., Parvez, F., 2017. Associations between prenatal arsenic exposure with adverse pregnancy outcome and child mortality. *Environ. Res.* 158, 456–461.
- Shrivastava, A., Barla, A., Singh, S., Mandraha, S., Bose, S., 2017. Arsenic contamination in agricultural soils of Bengal deltaic region of West Bengal and its higher assimilation in monsoon rice. *J. Hazard Mater.* 324, 526–534.
- Singh, R., Singh, S., Parihar, P., Singh, V.P., Prasad, S.M., 2015. Arsenic contamination, consequences and remediation techniques: a review. *Ecotoxicol. Environ. Saf.* 112, 247–270.
- Smedley, P., 2005. Arsenic Occurrence in Groundwater in South and East Asia.
- Smedley, P., Kinniburgh, D., 2002. A review of the source, behaviour and distribution of arsenic in natural waters. *Appl. Geochem.* 17, 517–568.
- Smith, A.H., Lingas, E.O., Rahman, M., 2000. Contamination of drinking-water by arsenic in Bangladesh: a public health emergency. *Bull. World Health Organ.* 78, 1093–1103.
- Sönmez, O., Turan, V., Kaya, C., 2016. The effects of sulfur, cattle, and poultry manure addition on soil phosphorus. *Turk. J. Agric. For.* 40 (4), 536–541. <https://doi.org/10.3906/tar-1601-41>.
- Stollenwerk, K.G., Breit, G.N., Welch, A.H., Yount, J.C., Whitney, J.W., Foster, A.L., Uddin, M.N., Majumder, R.K., Ahmed, N., 2007. Arsenic attenuation by oxidized aquifer sediments in Bangladesh. *Sci. Total Environ.* 379, 133–150.
- Su, C., Zhu, Y., Abbas, Z., Huq, M.E., 2016. Sources and controls for elevated arsenic concentrations in groundwater of Datong Basin, Northern China. *Environ. Earth Sci.* 75, 1–13.
- Tanvir Rahman, M.A.T.M., Saadat, A.H.M., Islam, M.S., Al-Mansur, M.A., Ahmed, S., 2017. Groundwater characterization and selection of suitable water type for irrigation in the western region of Bangladesh. *Appl. Water Sci.* 7, 233–243.
- Tapase, S.R., Kodam, K.M., 2018. Assessment of arsenic oxidation potential of *Microvirga indica* S-M11b sp. nov. in heavy metal polluted environment. *Chemosphere* 195, 1–10.
- Tareq, S.M., Maruo, M., Ohta, K., 2013. Characteristics and role of groundwater dissolved organic matter on arsenic mobilization and poisoning in Bangladesh. *Phys. Chem. Earth* 58, 77–84.
- Tareq, S.M., Safiullah, S., Anwar, H., Rahman, M.M., Ishizuka, T., 2003. Arsenic pollution in groundwater: a self-organizing complex geochemical process in the deltaic sedimentary environment, Bangladesh. *Sci. Total Environ.* 313, 213–226.
- Tuinhof, A., Kemper, K., 2010. Mitigation of Arsenic Contamination in Drinking Watersupplies of Bangladesh-the Case of Chapai Nawabganj. World Bank.
- Turan, V., Khan, S.A., Mahmood-ur, R., Ramzani, P.M.A., Fatima, M., 2018. Promoting the productivity and quality of brinjal aligned with heavy metals immobilization in a wastewater irrigated heavy metal polluted soil with biochar and chitosan. *Ecotoxicol. Environ. Saf.* 161, 409–419. <https://doi.org/10.1016/j.ecoenv.2018.05.082>.
- Turan, V., Ramzani, P.M.A., Ali, Q., Irum, A., Khan, W.-U.-D., 2017. Alleviation of nickel toxicity and an improvement in zinc bioavailability in sunflower seed with chitosan and biochar application in pH adjusted nickel contaminated soil. *Arch. Agron Soil Sci.* 64 (8), 1053–1067. <https://doi.org/10.1080/03650340.2017.1410542>.
- UNICEF, 2007. Predicting the Global Extent of Arsenic Pollution of Groundwater and its Potential Impact on Human Health, New York. Report Prepared by Peter Ravenscroft, Consultant Hydrogeologist, Cambridge, UK.
- van Geen, A., Ahmed, E.B., Pitcher, L., Mey, J.L., Ahsan, H., Graziano, J.H., Ahmed, K.M., 2014. Comparison of two blanket surveys of arsenic in tubewells conducted 12years apart in a 25km² area of Bangladesh. *Sci. Total Environ.* 488, 484–492.
- van Geen, A., Zheng, Y., Cheng, Z., He, Y., Dhar, R., Garnier, J., Rose, J., Seddique, A., Hoque, M., Ahmed, K., 2006. Impact of irrigating rice paddies with groundwater containing arsenic in Bangladesh. *Sci. Total Environ.* 367, 769–777.
- Verma, S., Mukherjee, A., Choudhury, R., Mahanta, C., 2015. Brahmaputra river basin groundwater: solute distribution, chemical evolution and arsenic occurrences in different geomorphic settings. *J. Hydrol. Reg. Stud.* 4, 131–153.
- von Brömsen, M., Markussen, L., Bhattacharya, P., Ahmed, K.M., Hossain, M., Jacks, G., Sracek, O., Thunvik, R., Hasan, M.A., Islam, M.M., 2014. Hydrogeological investigation for assessment of the sustainability of low-arsenic aquifers as a safe drinking water source in regions with high-arsenic groundwater in Matlab, southeastern Bangladesh. *J. Hydrol* 518, 373–392.
- Wang, Y., Jiao, J.J., 2014. Multivariate statistical analyses on the enrichment of arsenic with different oxidation states in the Quaternary sediments of the Pearl River Delta, China. *J. Geochem. Explor.* 138, 72–80.
- Wang, Y., Pi, K., Fendorf, S., Deng, Y., Xie, X., 2019. Sedimentogenesis and hydrobiogeochemistry of high arsenic Late Pleistocene-Holocene aquifer systems. *Earth Sci. Rev.* 189, 79–98.
- Wang, Y., Shpeyzer, G., 2000. Hydrogeochemistry of Mineral Waters from Rift Systems on the East Asia Continent: Case Studies in Shanxi and Baikal. China Environmental Science Press, Beijing (in Chinese with English abstract).
- Wang, Y., Shvartsev, S.L., Su, C., 2009. Genesis of arsenic/fluoride-enriched soda water: a case study at Datong, northern China. *Appl. Geochem.* 24, 641–649.
- Wang, Z., Ma, T., Zhu, Y., Liu, L., 2017. Removing arsenic from groundwater using a single well based on composite filtration materials comprised with siderite and sands. *Proced. Earth Plan. Sci.* 17, 400–403.
- Welch, A.H., Westjohn, D., Helsel, D.R., Wanty, R.B., 2000. Arsenic in ground water of the United States: occurrence and geochemistry. *Ground Water* 38, 589–604.
- Whaley-Martin, K., Mailloux, B., van Geen, A., Bostick, B., Ahmed, K., Choudhury, I., Slater, G., 2017. Human and livestock waste as a reduced carbon source contributing to the release of arsenic to shallow Bangladesh groundwater. *Sci. Total Environ.* 595, 63–71.
- WHO, 2011. Guidelines for Drinking-Water Quality, fourth ed.
- World Bank, 2018. Promising Progress: A Diagnostic of Water Supply, Sanitation, Hygiene, and Poverty in Bangladesh. WASH Poverty Diagnostic, World Bank Washington, DC.
- Xiao, T., Dai, Z., McPherson, B., Viswanathan, H., Jia, W., 2017. Reactive transport modeling of arsenic mobilization in shallow groundwater: impacts of CO₂ and brine leakage. *Geomec. Geophys. Geo-Ener. Geo-Resour.* 3, 339–350.
- Xie, X., Wang, Y., Ellis, A., Liu, C., Duan, M., Li, J., 2014. Impact of sedimentary provenance and weathering on arsenic distribution in aquifers of the Datong basin, China: constraints from elemental geochemistry. *J. Hydrol* 519, 3541–3549.
- Xie, X., Wang, Y., Su, C., 2012a. Hydrochemical and sediment biomarker evidence of the impact of organic matter biodegradation on arsenic mobilization in shallow aquifers of Datong Basin, China. *Water Air Soil Pollut.* 223, 483–498.
- Xie, X., Wang, Y., Su, C., Li, J., Li, M., 2012b. Influence of irrigation practices on arsenic mobilization: evidence from isotope composition and Cl/Br ratios in groundwater from Datong Basin, northern China. *J. Hydrol* 424, 37–47.
- Xu, X., Chen, C., Wang, P., Kretzschmar, R., Zhao, F.-J., 2017. Control of arsenic mobilization in paddy soils by manganese and iron oxides. *Environ. Pollut.* 231, 37–47.
- Yaghi, N., Hartikainen, H., 2018. Effect of oxide coatings, pH and competing anion on the sorption of arsenic species onto Light Expanded Clay Aggregates (LECA's). *Environ. Technol. Innov.* 9, 30–37.
- Yang, G.M., Zhu, L.J., Santos, J.A., Chen, Y., Li, G., Guan, D.X., 2017. Effect of phosphate minerals on phytoremediation of arsenic contaminated groundwater using an arsenic-hyperaccumulator. *Environ. Technol. Innov.* 8, 366–372.
- Yu, Q., Wang, Y., Xie, X., Currell, M., Pi, K., Yu, M., 2015. Effects of short-term flooding on arsenic transport in groundwater system: a case study of the Datong Basin. *J. Geochem. Explor.* 158, 1–9.
- Yu, Y., Guo, Y., Zhang, J., Xie, J., Zhu, Y., Yan, J., Wang, B., Li, Z., 2017. A perspective of chronic low exposure of arsenic on non-working women: risk of hypertension. *Sci. Total Environ.* 580, 69–73.
- Zahida, Z., Hafiz, F.B., Zulfikar, A.S., Ghulam, M.S., Fahad, S., Muhammad, R.A., Hafiz, M.H., Wajid, N., Muhammad, S., 2017. Effect of water management and silicon on germination, growth, phosphorus and arsenic uptake in rice. *Ecotoxicol. Environ. Saf.* 144, 11–18.

- Zhang, J., Ma, T., Feng, L., Yan, Y., Abass, O.K., Wang, Z., Cai, H., 2017a. Arsenic behavior in different biogeochemical zonation approximately along the groundwater flow path in Datong Basin, northern China. *Sci. Total Environ.* 584, 458–468.
- Zhang, L., Qin, X., Tang, J., Liu, W., Yang, H., 2017b. Review of arsenic geochemical characteristics and its significance on arsenic pollution studies in karst groundwater, Southwest China. *Appl. Geochem.* 77, 80–88.
- Zhang, P.P., Lyu, S.S., Zhu, X.H., Chen, X.-G., Wu, D.-D., Ye, Y., 2015. Assessment of arsenic contamination in and around a plateau lake: influences of groundwater and anthropogenic pollution. *Pol. J. Environ. Stud.* 24, 2715–2725.
- Zhang, Y., Yang, M., Huang, X., 2003. Arsenic(V) removal with a Ce(IV)-doped iron oxide adsorbent. *Chemosphere* 51, 945–952.
- Zhang, Z., Moon, H.S., Myneni, S.C., Jaffé, P.R., 2017c. Phosphate enhanced abiotic and biotic arsenic mobilization in the wetland rhizosphere. *Chemosphere* 187, 130–139.
- Zhao, F.-J., McGrath, S.P., Meharg, A.A., 2010. Arsenic as a food chain contaminant: mechanisms of plant uptake and metabolism and mitigation strategies. *Annu. Rev. Plant Biol.* 61, 535–559.
- Zhu, N., Qiao, J., Yan, T., 2019. Arsenic immobilization through regulated ferolysis in paddy field amendment with bismuth impregnated biochar. *Sci. Total Environ.* 648, 993–1001.