

## Research article

# High arsenic contamination and presence of other trace metals in drinking water of Kushtia district, Bangladesh



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## ABSTRACT

Drinking water with excessive concentration levels of arsenic (As) is a great threat to human health. A hydrochemical approach was employed in 50 drinking water samples (collected from Kushtia district, Bangladesh) to examine the occurrence of geogenic As and the presence of trace metals (TMs), as well as the factors controlling As release in aquifers. The results reveal that the drinking water of shallow aquifers is highly contaminated by As (6.05–590.7 µg/L); 82% of samples were found to exceed the WHO recommended limit (10 µg/L) for potable water, but the concentrations of Si, B, Mn, Sr, Se, Ba, Fe, Cd, Pb, F, U, Ni, Li, and Cr were within safe limits. The Ca-HCO<sub>3</sub>-type drinking water was identified as having high contents of As, pH and HCO<sub>3</sub><sup>-</sup>, a medium-high content EC, and low concentrations of NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup>, and Cl<sup>-</sup>. The significant correlation between As and NO<sub>3</sub><sup>-</sup> indicates that NO<sub>3</sub><sup>-</sup> might be attributed to the use of phosphate fertilizers and a factor responsible for enhancing As in aquifers. The study also reports that the occurrence of high As and the presence of TMs in drinking water may be a result of local anthropogenic activities, such as irrigation, intensive land use and the application of agrochemicals. The insignificant correlation between As and SO<sub>4</sub><sup>2-</sup> demonstrated that As is released from SO<sub>4</sub><sup>2-</sup> minerals under reducing conditions. An elevated pH value along with decoupling of As and HCO<sub>3</sub><sup>-</sup> plays a vital role in mobilizing As to aquifer systems. Moreover, the positive relationship between As and Si indicated that As is transported in the biogeochemical environment. The reductive suspension of Mn(IV)-oxyhydroxides also accelerated the As mobilization process. Over exploitation of tube-well water and the competitive ion exchange process are also responsible for the release of As in aquifers.

## 1. Introduction

Groundwater of hand tube-wells is the major source of potable water in Bangladesh (Majumder and Banik, 2019; Tareq et al., 2013). However, a severe public health crisis has emerged due to the prevalent threat of geogenic arsenic (As) in tube-well water. It is creating risks of different diseases, such as cancer, melanosis, hyperkeratosis, and black foot disease (Islam et al., 2017a; Rahman et al., 2018). Often, it is regarded as the silent toxin, since the adverse impacts of As poisoning are only apparent after a long period. In addition, elevated level of trace metals (TMs), such as Si, B, Mn, Sr, Ba, Fe, Cd, Pb, F, U, Ni, Li, and Cr, as

well as the low concentrations of elements such as Sb and Se, might amplify the toxicity of As (Goodsell et al., 2017; Islam et al., 2019; Lu et al., 2018; Pham et al., 2017). The people living in As-contaminated regions are vulnerable to high As and multi-metal concentrations. This situation can create different critical health problems. Many countries around the world have been identified where As concentrations in drinking water are above the World Health Organization (WHO) recommended value of 10 µg/L for drinking water (WHO, 2011). Particularly, certain Asian countries, namely, Bangladesh, China, Cambodia, India, Indonesia, Myanmar, Mongolia, Nepal, Pakistan, Thailand, and Vietnam, are well known for problems with As contamination (Gomez-

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Gonzalez et al., 2016; Hill et al., 2009; Huq et al., 2018b; Kanel et al., 2013; Kippler et al., 2016; Kumar et al., 2016; Rasool et al., 2017; Su et al., 2016).

Asian countries have several similar hydrogeological features, such as alluvial areas with low-lying flat topography, Holocene sediments that originated from big rivers, the existence of degradable organic matter, slow-moving groundwater, etc. (Lee et al., 2018; Ratna Reddy et al., 2017). These hydrogeological environments are considered to be the major causes for the natural occurrence of As in the groundwater (Chakraborty et al., 2015). The Bengal Delta is a recent floodplain and is reported as having high As contamination. Groundwater can be contaminated either by natural (geochemical weathering) and/or by anthropogenic causes (discharging of unrefined wastewater) (Bibi et al., 2008). In the aquifers of Bangladesh, As is mobilized both naturally (for instance, rock weathering) and artificially (such as through mining, irrigation and over exploitation of groundwater) (Islam et al., 2017b; Martin et al., 2015; Mukherjee et al., 2017). However, TM pollution in groundwater as well as drinking water is a key concern worldwide because of their persistence, rapid expansion, and chronic toxicity in the groundwater environment (Costa et al., 2018; Ersbøll et al., 2018; He et al., 2018; Pietrelli et al., 2019; Uppal et al., 2019). The presence of TMs in the environment is a result of anthropogenic inputs, and thus, they enter into subsurface sediments and groundwater. The anthropogenic sources of TMs include industrial waste effluence and the wide application of agrochemicals. The hydrochemical weathering of bedrock causes the natural occurrence of TMs and creates metallic pollution in the groundwater environment (Mukherjee et al., 2017; Rasool et al., 2016).

Globally, the dynamics of As and TMs have been investigated broadly in various ecosystems as well as the groundwater environment in the past two decades. Research on the natural occurrence of As and other TMs in drinking water and their consequences for human health has been conducted in some Asian countries and in Bangladesh too. For instance, Rahman et al. (2016a) have shown the geochemical controls on the occurrence and distribution of As in the groundwater of Manikganj district, Bangladesh. Similarly, Islam et al. (2017b) revealed the spatial distribution of TMs in the groundwater of Rangpur district, Bangladesh. Previously, Ahsan et al. (2009) 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 drinking water of the country is an issue of major concern. These studies focused on the distribution of As and TMs in groundwater systems, where they exhibit distinct pattern in various areas in Bangladesh. Nevertheless, the cooccurrence of As and TMs in drinking water and the mechanisms of As release have not been explored in detail, particularly in Kushtia district, Bangladesh. However, the concentration of As in drinking water of this district is higher than the WHO recommended limit (Rahman and Rahaman, 2018; Rahman et al., 2018). Therefore, it is essential to identify the key processes and controlling factors of As pollution, which might assist in determining the pollution-free drinking water sources in this area. Therefore, the current study attempts to assess the drinking water quality, physiochemical characteristics and the association between As distribution and TMs in drinking water. The specific objectives of this study are: (i) to study the content of As and TMs in drinking water, (ii) to identify the main sources of the geogenic As that occurs in drinking water, and (iii) to understand the mechanisms and/or processes that are responsible for the release of As in the aquifers. This investigation would help local residents have drinking water free of As and other harmful TMs.

## 2. Materials and methods

### 2.1. Study area

A total of 12 villages (Fig. 1) in the Kushtia district in Bangladesh were selected as the geographical area for this study. The river Padma

and its tributaries (the Mathabhanga and the Hishna), local lakes, canals, ponds, and rainwater are the main sources of the surface water system of the study area. During the rainy season, the River Padma acts as an excellent drainage channel, with a large volume of surface water from this area (Haque et al., 2016). Based on the subsurface lithology and the water table, three kinds of aquifers are observed in the study area. First, the composite aquifer, which spreads up to 50 m (m) below ground level (bgl); second, the main aquifer that occurs in between 50 and 150 m bgl; and third, the deep aquifer, which exists at or above 200 m bgl (Haque et al., 2015). Table 1 presents the climatic and geological characteristics of the study area.

In total, 50 drinking water samples were randomly taken from different hand tube-wells and electrically driven pumps of the study area with depths between 20 and 80 m. Geographical locations of the sampling sites were recorded by a hand GPS (GARMIN 12™) device. Polyethylene bottles were used to collect the drinking water samples. Before collecting the samples, each bottle was washed carefully and rinsed by the specific tube-well and electric pump water. Each of the hand tube-wells or electric pumps was purged for approximately 5 min before taking the samples to avoid additional floating solids from the pipe and to make sure actual aquifer water was collected.

### 2.2. In situ measurements

A 0.45 μm fiber filter was used to filter all the drinking water samples at the sampling sites. Three strained subsamples were collected in prewashed 50 mL polyethylene bottles separately to analyze anions, cations as well as TMs. These subsamples were then preserved by 10% ultra-pure HNO<sub>3</sub>. The physiochemical properties, such as temperature (T), pH, total dissolved solid (TDS), and electrical conductivity (EC), were measured in situ with calibrated hand Hanna meters in the sampling period. Finally, all the bottles with sampled water were sealed with screw caps and preserved in a refrigerator at 4 °C for further analysis.

### 2.3. Chemical analytical methods

Bicarbonate (HCO<sub>3</sub><sup>-</sup>) was measured by the titration method within 24 h of sampling (Yu et al., 2015). All the chemical analyses (major anions, cations, and TMs) were completed at the State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences (Wuhan), China. An Ion Chromatograph (IC) (Metrohm-761 Compact IC) was used to measure the concentrations of major anions (e.g., SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, and NO<sub>3</sub><sup>-</sup>). Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) was used to determine the contents of major cations (Ca<sup>2+</sup>, Na<sup>+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup>). The total solute As concentration was measured using Hydride Generation-Atomic Fluorescence Spectrometry (HG-AFS) (Titan AFS 830). Inductively Coupled Plasma Mass Spectrometry (Agilent 7500ce ICP-MS) along with an auto sampler were employed to measure the concentrations of TMs (B, Mn, Be, Ba, Cd, Li, Fe, Se, Cr, Cu, Sb, Ni, Pb, Si, Co, Sr, V, Ti, and Zn) in the drinking water samples. Prior to the measurement of chemical properties of the samples all the instruments had undergone for 30 min to optimize the procedure. The typical analytical error for cations, anions, As, and TMs measurements were kept within ± 5%. The quality assurance for the key ion analyses were ensured by estimating the charge balance error. The charge equilibrium error of drinking water samples was less than 5% (QA-QC).

### 2.4. Statistical analyses and graphical tools

The Statistical Package for Social Sciences (SPSS) (version 20) was used for the statistical analyses, such as maximums, minimums, means, and analytical error. It was also applied to calculate the correlation matrix. The correlation matrix can determine the relationship among the concentration of total As, TMs, and other water properties. The

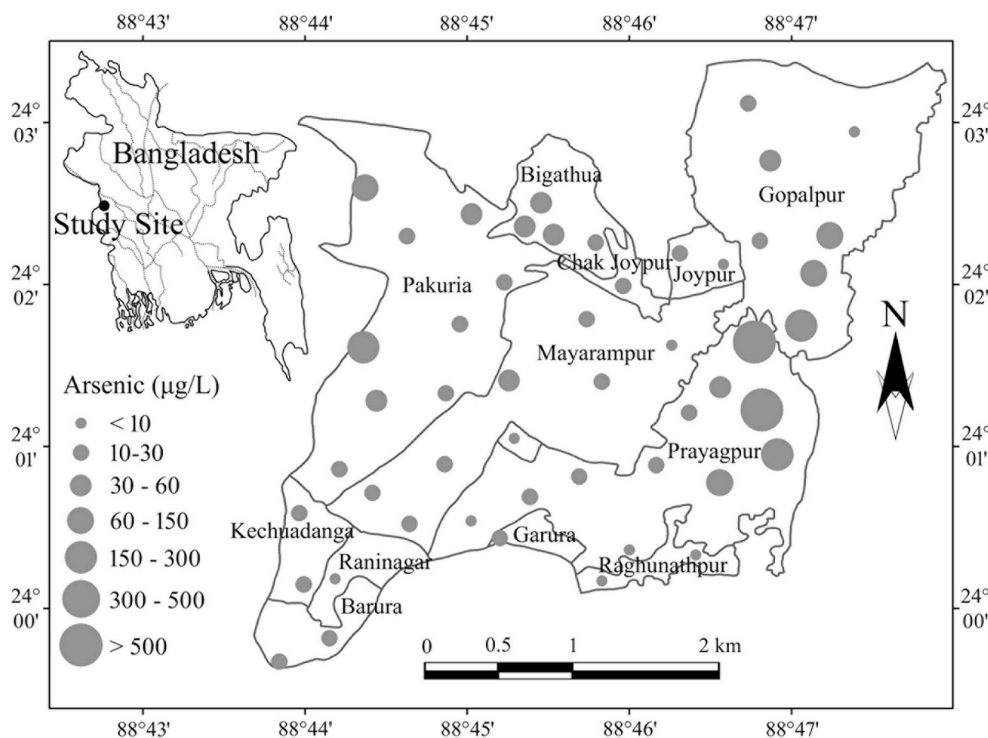


Fig. 1. Map of sampling locations with As concentration in drinking water of the study area.

Table 1

Climatic and geological characteristics of the study area.

Climatic Characteristics		Geological Characteristics	
Parameters	Description	Parameters	Description
Climate type	Subtropical monsoon climate	Surface geology	The surface geology is alluvium type, composed of clay, silt and fine sand, and gray-brown in color.
Major seasons	There are three major seasons (pre-monsoon, monsoon, and winter) (Rahman et al., 2014)	Components of soil	A mixture of clays, quartz, calcium carbonate, dolomite, mica, etc. The top clay and very fine sand layers exist everywhere.
Air temperature	The air temperature varies from 9 °C to 38.5 °C and the maximum temperature (38.5 °C) is recorded in April and May (BBS, 2015; Rahman and Rahaman, 2018).	Sediment type	Silt-medium sand and coarse-fine sand. Pleistocene older alluvium and Holocene sediments (Hossain et al., 2013).
Humidity	The average monthly relative humidity ranges in between 60% and 88%. The highest and lowest humidity are observed in July and March, respectively (Shahid and Khairulmaini, 2009).	Origin of sediment	The sediments mostly consist of quaternary deltaic deposits of rivers along with alluvial weathered sediments of the Himalayas.
Evaporation	The mean monthly maximum and minimum evaporation levels are 110 and 40 mm, respectively (Adhikary et al., 2012).	Slope and altitude	Almost a plain with gentle slope from north to south. The altitude ranges from 8 to 14 m above mean sea level (Haque et al., 2016).

AqQA RockWare software (version 1.1.1 [1.1.5.1] AqQA LLC, 2006) was applied to produce the Piper diagram to define the water type and assess the water chemistry of the drinking water. The box and whisker plots of major ions and TMs were prepared by OriginLab (version 8E, OriginLab Corporation, 2007). ArcGIS (version 10.2.2) was employed to draw the spatial distribution maps of As.

### 3. Results

#### 3.1. pH, EC, TDS and T

To investigate the drinking water quality, the physical and geochemical characteristics of the drinking water samples were measured. All the physical and geochemical data of drinking water samples are presented in Supplementary Material Table S1. The value of pH was a little alkaline (varies from 6.8 to 8.4, with an average of 7.25). The samples revealed moderate-high EC values (637-1280 µS/cm; average 989.5 µS/cm) with significant variation. A concentration of Total

Dissolved Solid (TDS) in drinking water above 1000 µg/L is intolerable for the human body, and a concentration of TDS of more than 2000 µg/L is unsafe for plants and crops (Sakizadeh, 2019). The TDS concentrations in the samples exhibited considerable variation (ranges from 173.6 to 509.7 mg/L with a mean of 260.1 mg/L). Temperatures of drinking water differed from 29 to 33.1 °C (Table S1). The average temperature was found to be 30.9 °C. The above findings of the present study are consistent with the previous studies of Ayers et al. (2016), and Reza et al. (2010a).

#### 3.2. Major ion chemistry

The concentrations of key ions such as Ca<sup>2+</sup> in drinking water samples varied from 6.66 to 86.06 mg/L, with an average of 26.49 mg/L (Table S1). The occurrence of Ca<sup>2+</sup> ions might be related to calcareous rocks as well as sediments (Ibrahim et al., 2016). Concentrations of Na<sup>+</sup> ranged from 6.53 to 40.44 mg/L. The Mg<sup>2+</sup> concentrations reached up to 31.94 mg/L, with a mean value of 23.38 mg/L in drinking

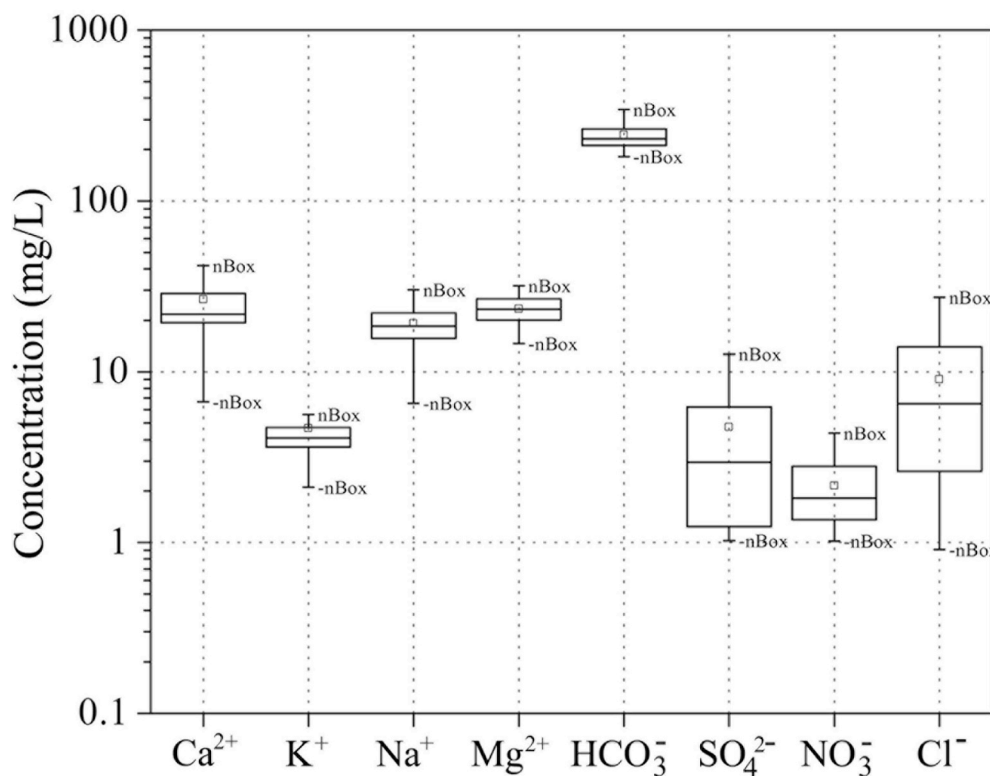


Fig. 2. Box and Whisker plot showing distribution of major ions.

water samples. Such results were also found by Radloff et al. (2017) and Rahman et al. (2016b). The minimum concentration of  $Mg^{2+}$  was 14.6 mg/L. The concentration of  $K^+$  ranged from 2.1 to 18.33 mg/L (Fig. 2). The presence of  $K^+$  was observed to be lower, in contrast to the other cations.

Considerable spatial variation of anions was noted for the drinking water of Kushtia district. The major anion was  $HCO_3^-$ , which ranged from 182.2 to 433.8 mg/L, with a mean value of 244.1 mg/L. The maximum  $HCO_3^-$  concentration (433.8 mg/L) was detected in the sample from DW 29 (Table S1). The  $NO_3^-$  concentration was not too high and varied from 1.02 to 6.74 mg/L in drinking water samples, but the concentrations of  $SO_4^{2-}$  were slightly higher (1.03–37.93 mg/L) than those of  $NO_3^-$  (Fig. 2). The  $Cl^-$  concentration varied significantly all over the investigated area, ranging from 0.9 to 44.25 mg/L. These analytical results are consistent with the prior studies of Howladar (2017) and Reza et al. (2010b). In this region, the chemistry of drinking water for  $K^+$  and  $SO_4^{2-}$  did not reveal any substantial variations. In Kushtia district, the  $Mg^{2+}$ ,  $Ca^{2+}$ , and  $HCO_3^-$  ions evolved from the dissolution of carbonates. In addition, dissolution of carbonate and high  $HCO_3^-$  normally originates from the degradation and oxidation of organic matter. Earlier studies (Huq et al., 2018a; Su et al., 2016) have noted that the elevated  $HCO_3^-$  plays a key role in hydrochemical evolution and TM mobilization in aquifers. Moreover, the chemical characteristics of the drinking water of this study are also comparable with the previous studies of BGS and DPHE (2001), Radloff et al. (2017), and Rahman et al. (2016b). Efforts were also made to find the mechanistic relationship of hydrogeochemistry using piper plots. Mainly, the Ca- $HCO_3^-$ -type drinking water was identified in this area (Fig. 3). The Ca- $HCO_3^-$ -type water is possibly resulted from the reductive dissolution of  $MnO(OH)$ , which is a factor that influences As release from alluvium to aquifers. The pressure of agricultural activities is also responsible for generating this type of water. Because, in the study area, for more production, different types of inorganic chemical fertilizer and pesticides are widely used. In addition, irrigation is very common in the cultivation of various types to cash crops and food grains. Mass water is

extracted for tobacco and rice cultivation.

### 3.3. As and other TMs

Concentrations of As and different TMs, including B, Mn, Be, Ba, Cd, Li, Fe, Se, Cr, Cu, Sb, Ni, Pb, Si, Co, Sr, V, Ti, and Zn, were also calibrated for the drinking water samples. The concentrations of As with other selected TMs in drinking water are shown in Table S1 and Fig. 4. Drinking water samples confirmed an elevated concentration of total As in the range 6.05–590.7  $\mu\text{g/L}$ , with an average of 58.31  $\mu\text{g/L}$ . The sites with major As concentrations are situated in the villages of Prayagpur, Pakuria and Bilgathua (Fig. 1). The maximum As (590.7  $\mu\text{g/L}$ ) contamination in drinking water was found in Prayagpur village. The concentrations of B, Ba, and F varied from 15.9 to 46.8  $\mu\text{g/L}$  (mean 27.51  $\mu\text{g/L}$ ), 12.8 to 208.6 (average 90.41  $\mu\text{g/L}$ )  $\mu\text{g/L}$ , and < 0.01–309.5  $\mu\text{g/L}$  (64.35  $\mu\text{g/L}$ ), respectively (Table S1 and Fig. 4). Research has shown that a small amount of B is essential for human health, sound bones, joint function, and for steroidal hormone metabolism (Hargreaves et al., 2018; Pokhrel and Viraraghavan, 2009; Rasool et al., 2016). In addition, the critical consequence of Ba on public health is still being determined.

On the other hand Se is a vital element for the human body that repels the cytotoxic effects of As (Chanpiwat et al., 2011). The Se concentrations in drinking water of Kushtia ranged from < 0.01 to 3.8  $\mu\text{g/L}$ , with a low mean value of 0.51  $\mu\text{g/L}$ . The low concentration of Se in drinking water could reduce the level of Se in the human body and thus increase the incidence of As-induced diseases. The occurrence of Ni was found between < 0.01 and 8.00  $\mu\text{g/L}$ , with an average value of 2.46  $\mu\text{g/L}$  (Table S1), which is higher than that of drinking water of the Datong Basin, China (0.94  $\mu\text{g/L}$ ) (Su et al., 2016). The current study found that the concentration of Cr (< 0.01–1.00  $\mu\text{g/L}$  with mean value 0.18  $\mu\text{g/L}$ ), V (< 0.01–1.5  $\mu\text{g/L}$  and an average of 0.19  $\mu\text{g/L}$ ), and Ti (< 0.01–0.6  $\mu\text{g/L}$  with the mean value 0.15  $\mu\text{g/L}$ ) were much lower than those of the other geochemical parameters. The concentration of Li (vary 2.8–22  $\mu\text{g/L}$  with a mean of 7.69  $\mu\text{g/L}$ ) was noticed to be

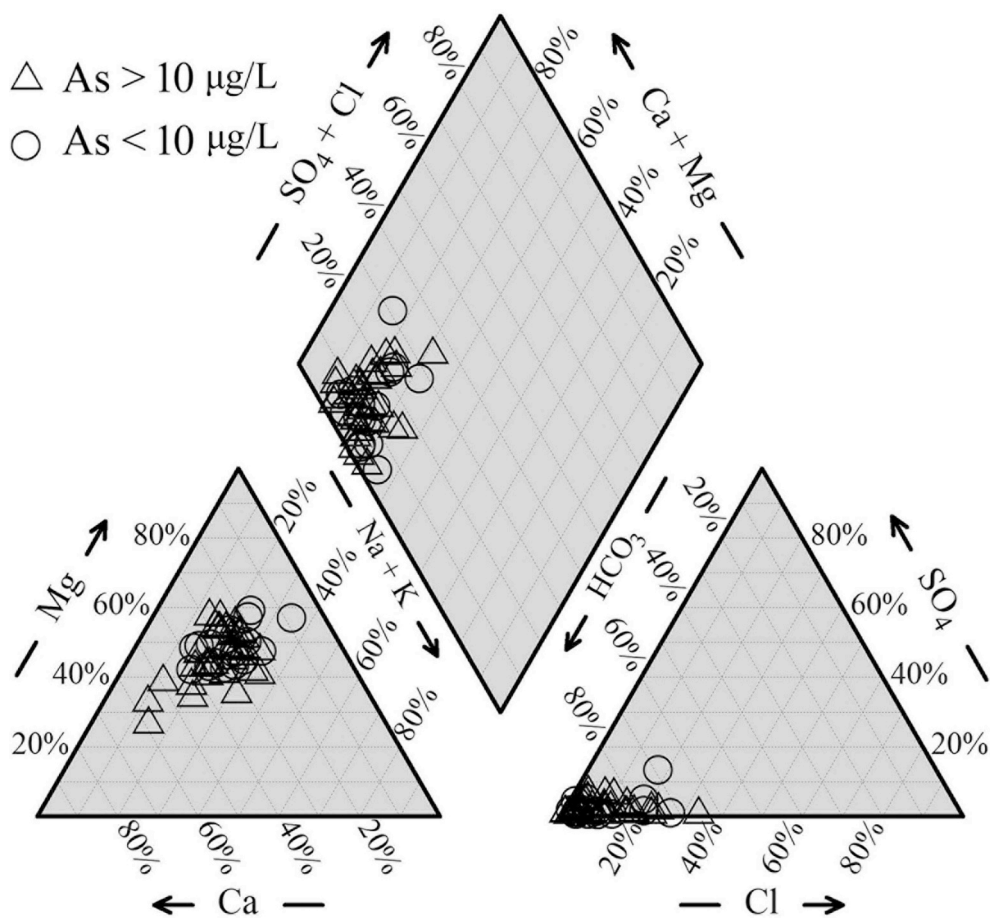


Fig. 3. Piper diagram of high-As drinking water of the study area.

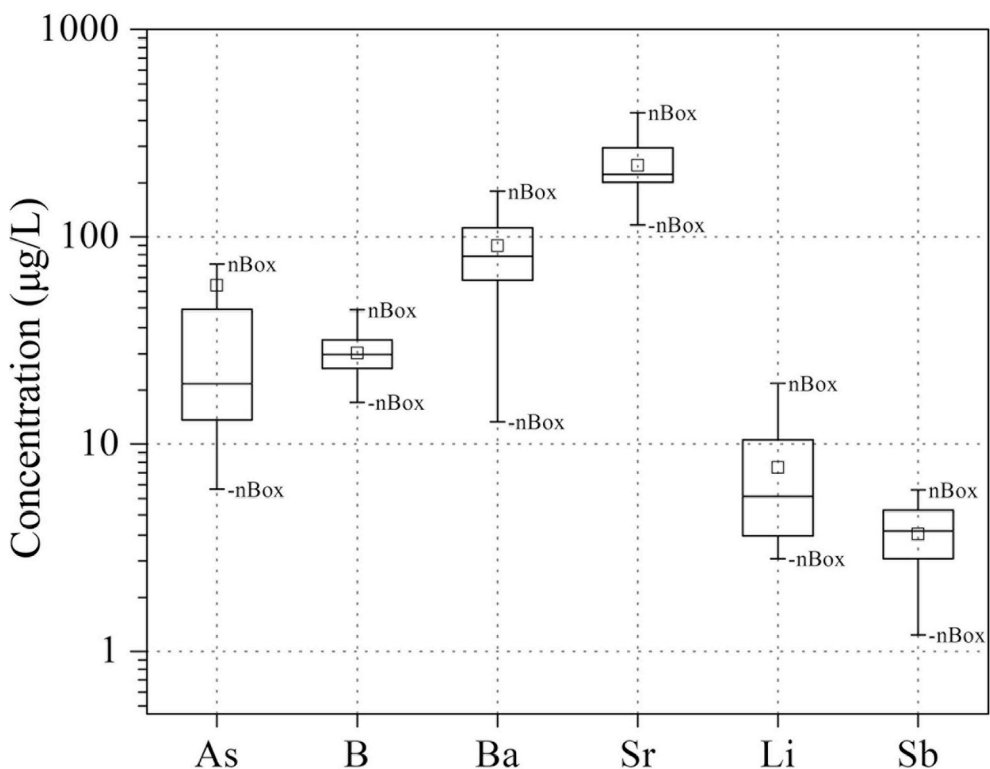


Fig. 4. Box and Whisker plot showing distribution of As and other trace metals.

comparatively elevated compared to the other TMs (Fig. 4).

However, Mn is a catalyst for several enzymatic processes that are related to the implication of fatty acids and cholesterol (Kumar et al., 2016). The concentrations of Mn in drinking water samples ranged from < 0.01 to 56.2 µg/L (average 1.4 µg/L). Moreover, some other TMs, including Pb, Sb, Si, Sr, Ti, and V, were also detected in drinking water samples. The concentrations of Pb, Sb, Ti, and V varied from 0.6 to 8.00 µg/L, 1.2–6.00 µg/L, < 0.01–0.6 µg/L, and < 0.01–1.5 µg/L, respectively (Table S1 and Fig. 4). The levels of Si and Sr ranged from 8.15 to 18.67 (mean 14.84 mg/L) and 0.026–0.403 mg/L (mean 0.221 mg/L), respectively (Fig. 4). The results mentioned above are consistent with the findings of Bodrud-Doza et al. (2016) and Howladar (2017). Elevated levels of As predominantly occurred in the drinking water in this area. Even the low concentrations of some TMs, such as Sb, Cr, Ti and V, might increase the toxic effect of As. Previous studies (Kumar et al., 2016; Mosler et al., 2010; Rahman et al., 2015; Shahmoradi et al., 2017) have demonstrated that the inhabitants living in As-polluted regions and consuming As-enriched water are exposed to elevated As concentrations and multiple TMs concentrations that could be the cause of adverse health impacts. The empirical results of this study confirm that the people of Kushtia district are vulnerable not only to As poisoning but also to contamination from B, Ba, F, Mn, and Sr, which can cause health hazards.

## 4. Discussion

### 4.1. As in groundwater

#### 4.1.1. Occurrence and spatial distribution

The occurrence of total dissolved As in drinking water of Kushtia district, Bangladesh was highly varied (ranging from 6.04 to 590.7 µg/L with an average value of 59.73 µg/L). Among the affected villages, Prayagpur, Gopalpur, Pakuria, and Bilgathua suffer most from As-contaminated tube-wells. Based on the spatial distribution, two As-contaminated hotspots were identified: (i) the area surrounding Prayagpur and Gopalpur villages, and (ii) Pakuria and Bilgathua villages (Fig. 5).

The As concentrations of these two hotspots reached up to 590.7 and 157.5 µg/L respectively. The unaffected or less As-affected regions are mostly situated in the middle of the studied site. The As-enriched shallow Holocene sand, resulting from the glacial weathering of the Himalaya serves as the main source of As in the Bengal Basin. In contrast, the deep Pleistocene rock-sediments derived from oxidative weathering act as the leading source of mobilizable As within the biogeochemical environment (Aderibigbe et al., 2018; Chakraborty et al., 2015). However, the suspended sediments of the surrounding rivers, irrigation return, and the recently deposited sediment contributed the high level of As in the soils of this district. During the monsoon floods, surface soil releases a huge volume of As to the floodwater, and subsequently, this As contaminates the shallow groundwater. The weathering of biotite might also significantly add As in aquifers of Kushtia district, which varies with depth.

#### 4.1.2. As concentration along with depth

The drinking water samples were collected from different depths (varying from 21 to 81 m). The As concentration and depth profile of drinking water samples revealed that the high As contamination in drinking water mostly occurred in the shallow aquifers ( $\leq 40$  m). Whereas the drinking water of deep aquifers ( $> 40$  m) contained less As. The sediment of this area is made up of clay, silt, fine and medium sand, very fine sand and coarse sand (Fig. 6a). The shallow aquifers fall in the depth ranging from 8 to 40 m and are reported to have elevated As concentrations. The result showed that the maximum concentration of As (590.7 µg/L) occurred at the depth of 33.5 m, and then it decreased with increasing of depth (Fig. 6b). Similar results were also found by Chakraborti et al. (2010) and Knappett et al. (2016). The shallow aquifers of Kushtia district consist of clay, very fine sand layers

and a thick layer of yellowish-brown silty clays that are covered by a thin layer of very fine sand and a thick clay layer at the top. This type of geological structure facilitates the occurrence of As (Chauhan et al., 2018; Ramos et al., 2014). The deep aquifer is formed by a coarse sand layer, which is underlain by another fine sandy deposit that continues up to 130 m (Fig. 6a). Moreover, the study area is a recent flood plain, which is comparatively loose, more fragile in nature and favorable to the release of As.

#### 4.1.3. Cooccurrence of As and salinity

Drinking water with high levels of TDS is harmful to the human body, plants and crops. Natural processes, such as strong evaporation, and human activities, for instance, intensive irrigation, as well as overexploitation of groundwater, are considered to be responsible for triggering and/or increasing the salinity in groundwater systems (Moradlou et al., 2016; Rahman et al., 2015). During the irrigation season, the groundwater table of this district goes down approximately 0.5–3 m compared to the non-irrigation season. The analyzed water samples showed the positive correlation (0.46) between TDS and EC values (Fig. 7b). This correlation indicates the salinity in groundwater (Chanpiwat et al., 2011; Jia et al., 2017). The high concentration of TDS (173.6–509.7 mg/L) in drinking water may be attributed to irrigation, residential use and dyeing units. As Tweed et al. (2018) demonstrated, the TDS concentrations prior to irrigation were lower than the TDS values during irrigation, which indicated an effect of irrigation on the occurrence of TDS. The elevated TDS levels of this area suggest the potential consequences of strong evapotranspiration, industrial toxic waste, and domestic sewage. The long-term water-rock reactions may lead to the occurrence of these types of TDS concentrations in reducing conditions (Chetia et al., 2012). The present study also found the co-existence of elevated As and salinity in the aquifer system of the study area, since analyzed samples confirmed a positive correlation (0.13) of As and TDS (Fig. 7a).

### 4.2. Role of ions and TMs in mobilizing As

Different hypotheses have been developed to explain the release and transportation process of As in groundwater (Pi et al., 2015). However, these hypotheses do not explain the process clearly. However, Harvey et al. (2006) noted that a negative correlation between As and  $\text{SO}_4^{2-}$  is observed in groundwater (which is using as drinking water) of Holocene aquifers in Bangladesh. The low concentrations (average 4.37 mg/L) of  $\text{SO}_4^{2-}$  in drinking water samples and inverse correlation ( $-0.12$ ) of As with  $\text{SO}_4^{2-}$  (Table 2) demonstrates that As is released from  $\text{SO}_4^{2-}$  minerals in the Kushtia district under reducing conditions. The As in  $\text{SO}_4^{2-}$  minerals are easily extractable and weakly sorbed. This sorbed As in  $\text{SO}_4^{2-}$  minerals might mobilize under oxidizing conditions (Chetia et al., 2012; Ramos et al., 2014). The existence of  $\text{SO}_4^{2-}$  in the hand pump tube-well water may be attributed to pit latrines. In the study area, the mean value of  $\text{SO}_4^{2-}$  in drinking water was low, but the clayey silts as well as sands comprise a significant amount of As (Fig. 6a). Groundwater As is also released by the competitive ion exchange of  $\text{NO}_3^-$ , which is attributed to fertilizer. In addition,  $\text{NO}_3^-$  is supposed to be a major key competitor of ion exchange with As. However, as the  $\text{HCO}_3^-$  ion displaces As from sediments, it has a significant role in the mobilization of As. Moreover, the As-enriched water containing high  $\text{HCO}_3^-$  releases sorbed As due to the competitive displacement of As (Michael and Khan, 2016). The strong positive correlation (0.48) of  $\text{Mg}^{2+}$  and  $\text{Na}^+$  (Table 2) suggests that  $\text{Mg}^{2+}$  and  $\text{Na}^+$  may come from the weathering of silicate or carbonate suspension and the dissolution of the soil salts. This result can be compared with the earlier study of Rahman et al. (2016b). Moreover, groundwater with excessive exchangeable  $\text{Mg}^{2+}$  hinders the infiltration process (Pi et al., 2016). Analytical results showed that the low content of  $\text{Mg}^{2+}$  (ranges from 14.6 to 31.94 mg/L) implies a favorable infiltration environment that facilitates the accessibility of surface As into the aquifer of the

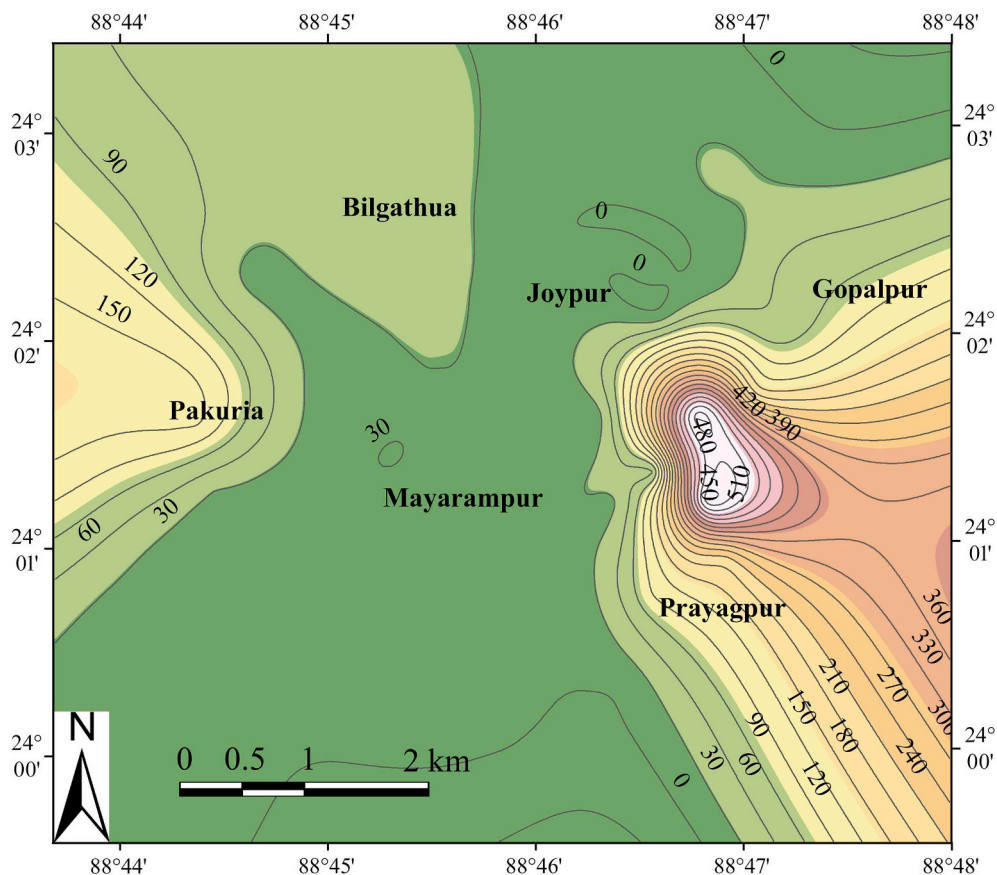


Fig. 5. Contour map of As concentration in drinking water of the study area.

study site. Furthermore, the surface wash As-contaminated water could easily pollute the 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. Since the late Pleistocene, a huge amount of coweathered As from sulfide minerals has been 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; Roy et al., 2015). The positive correlation (0.14) between As and Si (Table 2) is presumed to be the possible indicator of As sorption and mobilized As in

groundwater under a favorable biogeochemical environment. In addition, very low (mean 1.4 µg/L) Mn concentrations were found in drinking water samples (Table S1). This low concentration of Mn in aquifer is probably resulted from the reductive dissolution of Mn(IV)-oxyhydroxides, which accelerates the As mobilization process in the groundwater environment. Higher Ba concentrations in groundwater are an indicator of the terrestrial sediment environment, but low concentrations of Ba (Table S1) and the inverse correlation of As and Ba (Table 2) of the samples suggest the inferiority of the terrestrial sedimentary groundwater environment. Apparently, this type of

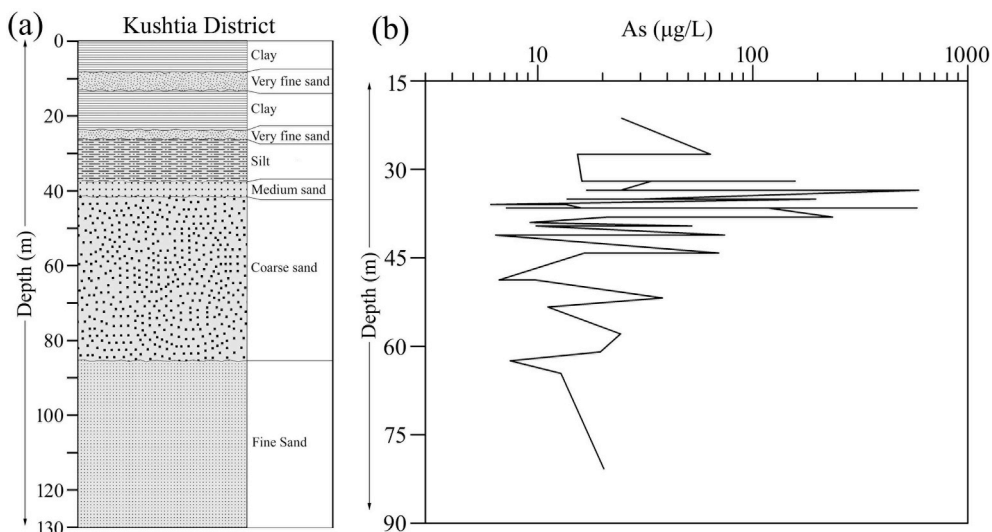


Fig. 6. (a) Geological section of Kushtia district (BGS and DPHE, 2001) and (b) Depth profile of As.

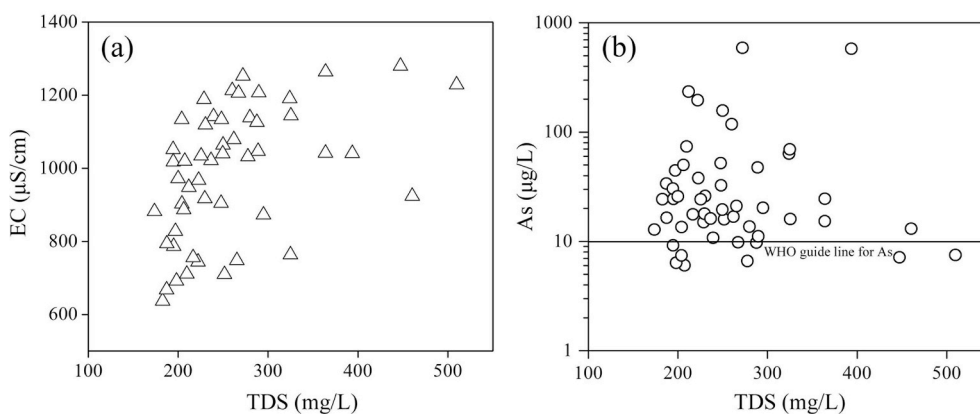


Fig. 7. Relationship between (a) EC and TDS (b) Arsenic and TDS.

environment facilitates the enrichment of As.

The poor correlation among As and other hydrochemical parameters (e.g., Ba and Mn) indicates that the mobilization of As is caused by multiple complex processes (Table 2). The concentration of Pb (0.6–8.0 µg/L) in drinking water was very low, which might be attributed to the precipitation of gasoline in the recent past. However, the determination of the sources of Pb in the drinking water of Kushtia district needs further investigation. The terrestrial sources of natural matter may be the geochemical factor responsible for the release of As in groundwater due to the high population density and intensive land use pattern of this area. The presence of cow dung in sediment of Bangladesh can increase soluble aqueous levels of As through binding with natural organic matter (Lin et al., 2017; Neumann et al., 2011). Moreover, use of local cow dung (easily degradable and decomposable by microbial action) during installation of tube-wells as the cementing material could also be the carbon source that can be a cause for high As concentrations in the newly installed tube-wells. Additionally, cow dung has notable fractions of ionically exchangeable functional groups, including carboxylic groups, which play an important role in As release. Utilization of natural organic matter-enriched cow dung fertilizer can promote As enrichment in aquifers (Kar et al., 2011; Majumder et al., 2014). However, the relationship between cow dung and As contamination has been discovered recently, and therefore, there is a need for more detailed study regarding this issue.

#### 4.3. Use of agrochemicals and its consequences in the study area

Most of the farmland in different districts of Bangladesh is threatened due to excessive use of agrochemicals (i.e., pesticides). Thus, the adverse impacts of agrochemicals on the food chain, drinking water and public health have increased (Biswas et al., 2014; Chowdhury et al., 2017; Polizzotto et al., 2013). Agrochemicals (pesticides and fertilizer) may affect the formation of groundwater as well as microbial elements

of aquifers through direct injection into groundwater. In addition, industrial discharges (Govindarajan and Senthilnathan, 2014) and municipal activities can influence the groundwater quality (Parveen and Nakagoshi, 2001). For example, Huq et al. (2018b) mentioned that the wastewater from chemical and mining industries and over-application of lime and gypsum in agricultural land also may be possible sources of contamination in groundwater of Kushtia district. Thus, the use of agrochemicals in cropland might affect the physical, chemical, and bacteriological characteristics of the surface water and groundwater. Moreover, the harmful and excessive use of agrochemicals can also pollute (such as pollution) the ground and surface water (Rahman and Rahaman, 2018). Uncontrolled application of fertilizers, pesticides and irrigation in rice fields caused sediment pollution. However, while farming soils are the primary receivers of agrochemicals, the surface water bodies (one of the recharge sources of groundwater) that are next to agricultural lands are typically the final recipient of agrochemical residues (Kutub and Falgunee, 2017; Mukherjee et al., 2017). Therefore, agrochemical residues are very common in surface water (e.g., ponds and rivers). It also harmful for the groundwater environment as well (Biswas et al., 2014). The As in levels in agricultural food crops can be high due to use of affluent As fertilizers (Ahmed et al., 2016). In the study area, different agricultural activities (such as farming of grains, cash crops and especially tobacco) are present, and for farming these crops, agrochemicals are used extensively (Kutub and Falgunee, 2017; Saha et al., 2016).

The farmers of Kushtia district use a large amount of agrochemical fertilizers (potassium, phosphorous as well as urea), pesticides (Aldicarb, Chlorpurifos and 1,3-dichloropropene), and irrigation to get maximum production, which causes air, water and soil pollution, the destruction of biodiversity, deforestation as well as other environmental degradation (Hossain and Rahman, 2013; Saha et al., 2016). 1,3-dichloropropene infiltrates into groundwater due to the application of the broad-spectrum soil fumigant to control nematodes of the soil.

Table 2

Pearson correlation matrix among As, other ions, and metals.

	As	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	Si	Ba	Mn
As	1							
SO <sub>4</sub> <sup>2-</sup>	-0.125229	1						
NO <sub>3</sub> <sup>-</sup>	0.392244 <sup>a</sup>	0.004679	1					
Mg <sup>2+</sup>	0.084587	0.270893 <sup>b</sup>	-0.073219	1				
Na <sup>+</sup>	-0.048516	0.533182 <sup>a</sup>	-0.056323	0.483265 <sup>a</sup>	1			
Si	0.143498	-0.405348 <sup>a</sup>	-0.124640	-0.222068	-0.104475	1		
Ba	-0.083510	0.422719 <sup>a</sup>	-0.064368	0.268973 <sup>b</sup>	0.247308 <sup>b</sup>	-0.216569	1	
Mn	-0.042416	0.016979	-0.125003	0.025027	0.066149	0.135298	-0.073423	1

<sup>a</sup> Correlation is significant at the 0.01 level (1-tailed).

<sup>b</sup> Correlation is significant at the 0.05 level (1-tailed).

The poisonous elements of aldicarb sulfoxide and aldicarb sulfonic also enter into aquifers and makes the groundwater toxic. The nitrate, phosphorous and dissolved oxygen in the soils of the study area mostly originated from agrochemical fertilizers (Biswas et al., 2014; Hossain and Rahman, 2013). These compounds pollute the groundwater and create eutrophication in the drinking water of the study site. Moreover, use of organic components, for instance, animal manures, biosolids, or composts, is the source of heavy metals in agricultural fields. The intensive agricultural practices with excessive use of agrochemicals in farmland in the Kushtia district has caused the complex accumulation of As and other TMs (Saha et al., 2016). Additionally, various extreme hazardous agrochemicals are being applied in Bangladesh, though they are prohibited in manufacturing countries. In addition, the existing agrochemical rules and principles are not accurately implemented in Bangladesh (Hill et al., 2009). Hence, to reduce the public health hazard, the rules and regulations on agrochemical abuse in agricultural land must be imposed, and continuous monitoring of As and TMs in soil and groundwater is needed.

#### 4.4. Drinking water quality with respect to WHO, USEPA and Bangladesh standards

The guidelines suggested by the WHO, United States Environmental Protection Agency (USEPA) and Bangladesh National Drinking Water Quality Survey for potable water with the quality of drinking water of the study area are presented in Table 3. Generally, the majority of drinking water samples had pH levels within the range of WHO guidelines and Bangladesh standards (6.5–8.5). However, 12% drinking water samples exceeded the USEPA standard (6–8) (USEPA, 2009). However, in accordance with the WHO and Bangladesh standards of drinking water, the quality of sampled drinking water ranged between ‘good’ and ‘poor’. Based on the taste, samples could be distinguished as good quality drinking water. None of the analyzed water samples crossed the EC value of WHO guidelines (1500  $\mu\text{S}/\text{cm}$ ). All the analyzed samples showed concentrations of TDS lower than the WHO and USEPA suggested value (1000 mg/L), as well as Bangladesh standard (Table 3). The concentrations of  $\text{Na}^+$  and  $\text{Mg}^{2+}$  of the drinking water samples of Kushtia district did not exceed the WHO acceptable limit, as well as the USEPA and Bangladesh standard. In general, the  $\text{K}^+$  was not greater than the WHO permissible level (30 mg/L) and USEPA standard (20 mg/L) for drinking water in the study area. However, the

**Table 3**

The quality of drinking water of the study area with WHO, USEPA and Bangladesh Standards.

Parameters	WHO guideline values	USEPA Standard	Bangladesh Standard	In the study area	
				Minimum	Maximum
pH	6.5–8.5	6–8	6.5–8.5	6.8	8.4
EC ( $\mu\text{S}/\text{cm}$ )	1500	–	–	637	1280
TDS (mg/L)	1000	2000	1000	173.6	509.7
$\text{Cl}^-$ (mg/L)	250	170	150–600	0.9	44.25
$\text{SO}_4^{2-}$ (mg/L)	250	250	400	1.03	37.93
$\text{NO}_3^-$ (mg/L)	50	30	10	1.02	6.74
$\text{Ca}^{2+}$ (mg/L)	200	70	75	6.66	86.06
$\text{K}^+$ (mg/L)	30	20	12	2.1	18.33
$\text{Mg}^{2+}$ (mg/L)	150	20	30–35	14.6	31.94
$\text{Na}^+$ (mg/L)	200	200	200	6.53	40.44
$\text{As}_{\text{total}}$ ( $\mu\text{g}/\text{L}$ )	10	10	50	6.05	590.7
B ( $\mu\text{g}/\text{L}$ )	500	400	1000	15.9	46.8
Ba ( $\mu\text{g}/\text{L}$ )	700	–	10	12.8	208.6
Cr ( $\mu\text{g}/\text{L}$ )	50	–	50	< 0.01	1.00
F ( $\mu\text{g}/\text{L}$ )	1500	–	1000	< 0.01	309.5
Mn ( $\mu\text{g}/\text{L}$ )	400	80	500	< 0.01	56.2
Ni ( $\mu\text{g}/\text{L}$ )	20	–	100	< 0.01	8.00
Pb ( $\mu\text{g}/\text{L}$ )	10	–	50	0.6	8.00
Sb ( $\mu\text{g}/\text{L}$ )	20	–	–	1.2	6.00
Se ( $\mu\text{g}/\text{L}$ )	10	–	10	< 0.01	3.8

concentration of  $\text{K}^+$  of two samples were more than the Bangladesh standard (12 mg/L). Moreover, the  $\text{Ca}^+$  concentration of a sample (DW29) showed a higher value than the USEPA and Bangladesh standard but was not more than the WHO recommended limit. Additionally,  $\text{Ca}^+$  does not play a typical role in adverse health impacts (BBS and UNICEF, 2011). According to the WHO guideline, the maximum permitted limit of the  $\text{SO}_4^{2-}$  ion in potable water is 250 mg/L. In this area, not a single drinking water sample exhibited a concentration of  $\text{SO}_4^{2-}$  above the WHO, USEPA and Bangladesh allowed limit. The other anions ( $\text{Cl}^-$  and  $\text{NO}_3^-$ ) revealed same results. However, the mean value of the As concentration (58.31  $\mu\text{g}/\text{L}$ ) of drinking water of Kushtia district was much higher than the WHO recommended limit and USEPA standard (10  $\mu\text{g}/\text{L}$ ). Even the average As concentration in drinking water of this area exceeded the Bangladesh standard (50  $\mu\text{g}/\text{L}$ ) for drinking water. The analytical results showed that 82% and 22% of drinking water samples contained As above the WHO suggested maximum value and the Bangladesh standard in potable water. The results showed that 41 samples out of the 50 contained As above the WHO suggested limit, and 11 samples exceeded the national limit for drinking water. In consideration of other TMs (excluding Ba), 100% of samples did not cross the WHO limit and the standard of USEPA and Bangladesh for drinking water (Table 3).

## 5. Conclusion

The present study demonstrates that there were several responsible sources for the high As contamination and occurrence of TMs in drinking water of Kushtia district, Bangladesh. Approximately 82% and 22% of drinking water samples contained As levels greater than the WHO (10  $\mu\text{g}/\text{L}$ ) and Bangladesh (50  $\mu\text{g}/\text{L}$ ) recommended values, respectively. Biotitic chemical weathering and the existence of other core minerals in Holocene aquifers are the primary sources of As in drinking water. Silicate weathering and degradation of organic matter are active processes in the study area and are the dominate geochemical processes that contaminate the shallow Holocene alluvial aquifers with As. However, the deep Holocene aquifers contain low dissolved As, but they are in a vulnerable condition owing to cross-contamination from the top shallow aquifers. The local geology, geomorphology, sediment types, groundwater flow, hydrochemical as well as anthropogenic factors, use of fertilizer, and adverse effects of irrigation control the occurrence of As in Kushtia district. Some chemical measures (oxidation of As(III) to As(V), coprecipitation of Fe/Mn oxidizing species and then filtration) and biological measures (determination of species of bacteria) can be taken to control As in drinking water. This study found that the  $\text{HCO}_3^-$  ion displaces As from sediments and mobilized sorbed As to the aquifers. Moreover,  $\text{SO}_4^{2-}$  plays a vital role in the release of As because elevated As in drinking water is found with strong reduction of  $\text{SO}_4^{2-}$ . The major ion chemistry of the study area was mostly influenced by evaporative rock-water interactions. The spatial variation of As concentration in this area exhibits considerable variability with geological characteristics. Furthermore, the analyzed geochemical data of drinking water samples suggest the following geochemical processes: (i) the existence of chemical weathering, (ii) an active As mobilization process, (iii) the release of As with the competitive ion exchange process, and (iv) the oxidation of sulfide minerals. Future research needs to be conducted to identify the mechanisms of the mobilization of other TMs in the aquifers.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2019.04.086>.

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