Environ Monit Assess (2011) 173:371–385
DOI 10.1007/s10661-010-1393-8

Groundwater arsenic contamination in Brahmaputra river basin: a water quality assessment in Golaghat (Assam), India

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Received: 30 July 2009 / Accepted: 11 February 2010 / Published online: 12 March 2010

Abstract Distribution of arsenic (As) and its compound and related toxicology are serious concerns nowadays. Millions of individuals worldwide are suffering from arsenic toxic effect due to drinking of As-contaminated groundwater. The Bengal delta plain, which is formed by the Ganga–Padma–Meghna–Brahmaputra river basin, covering several districts of West Bengal, India, and Bangladesh is considered as the worst As-affected alluvial basin. The present study was carried out to examine As contamination in the state of Assam, an adjoining region of the West Bengal and Bangladesh borders. Two hundred twenty-two groundwater samples were collected from shallow and deep tubewells of six blocks of Golaghat district (Assam). Along with total As, examination of concentration levels of other key parameters, viz., Fe, Mn, Ca, Na, K, and Mg with pH, total hardness, and SO_4^{2-}, was also carried out. In respect to the permissible limit formulated by the World Health Organization (WHO; As 0.01 ppm, Fe 1.0 ppm, and Mn 0.3 ppm for potable water), the present study showed that out of the 222 groundwater samples, 67%, 76.4%, and 28.5% were found contaminated with higher metal contents (for total As, Fe, and Mn, respectively). The most badly affected area was the Gamariguri block, where 100% of the samples had As and Fe concentrations above the WHO drinking water guideline values. In this block, the highest As and Fe concentrations were recorded 0.128 and 5.9 ppm, respectively. Tubewell water of depth 180 ± 10 ft found to be more contaminated by As and Fe with 78% and 83% of the samples were tainted with higher concentration of such toxic metals, respectively. A strong significant correlation was observed between As and Fe (0.697 at $p < 0.01$), suggesting a possible reductive dissolution of As–Fe-bearing minerals for the mobilization of As in the groundwater of the region.

Keywords Arsenic · Groundwater · Alluvium · Brahmaputra river basin

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Introduction

Natural potable water resource is becoming exceedingly a limited reserve for the human race throughout the world. Furthermore, presence of varied numbers of pollutants including heavy metals in the water through natural and/or anthropogenic interventions imparts toxic and harmful effects to the environment and the individual (Chatterjee et al. 2008; Vaclavikova et al. 2008). Among the pollutants, arsenic is considered a high-priority toxic metal because it acts as a human carcinogen (Jie and Waalkes 2008; Das et al. 2009; Henke 2009). The introduction of a redox-sensitive element like arsenic (standard reduction potential $E^\circ = 0.56$ V and standard oxidation potential $E^\circ = -0.67$ V) in the environment may take place through several ways. Eroded sediments and varied inputs of human activities like mining, pesticides, pharmaceuticals, etc. are thought to be the common sources of arsenic (Bunnell et al. 2007). Chronic arsenic exposure is detrimental to human health being associated with cancer of the skin, lung, liver, urinary bladder, and kidney (Tchounwou et al. 2003; Bunnell et al. 2007; Jie and Waalkes 2008) and other diseases, including cardiovascular and peripheral vascular diseases, diabetes, peripheral neuropathies, portal fibrosis, and adverse birth outcomes (Xia et al. 2009). The arsenic contamination of drinking water is among the most awesome environmental health challenges nowadays, threatening the well-being and livelihood of more than a hundred million people worldwide (Bhattacharya et al. 2007). The presence of arsenic in groundwater has been reported extensively in recent years from different parts of the world, including countries in North America and Latin America (viz., USA, Canada, Mexico, Argentina, Bolivia, Brazil, and Nicaragua), Australia, and Southeast Asia (viz., Bangladesh, China, Nepal, Vietnam, Cambodia, and India; Smedley and Kinniburgh 2002; Bhattacharya et al. 2006; Mukherjee et al. 2006; Acharyya and Shah 2007; Das et al. 2009). However, the environmental problem of arsenic toxicity in groundwater of the entire Bengal delta of the Ganga–Padma–Meghna–Brahmaputra (GPMB) river plain, covering several districts of West Bengal and Bangladesh, creates apprehension toward the scientific community and considered as the worst arsenic-affected alluvial basin (Smith et al. 2000; Das et al. 2009). The magnitude of the severity in respect with arsenic toxicity in the region, where subsurface water is primarily used as a source of drinking water, is very high. In Bangladesh, 28–77 million people drink arsenic-contaminated water without having alternative resources. Chronic consumption of such toxic water, in the future, may lead to, according to the estimate of the World Health Organization (WHO), 1 in every 10 adult deaths caused by arsenic-related cancer (Ahmad et al. 2003; Halem et al. 2009, SOS-Arsenic, downloaded from http://www.sos-arsenic.net on 28 Feb 2010). Again, the permissible limit for As in drinking water, according to the WHO, is 0.01 ppm, which is similar to the specification laid down by the Bureau of Indian Standards (BIS; BIS 1991; WHO 1993; Nickson et al. 2007); however, in India, according to the BIS (1991), the maximum permissible limit in the absence of an alternate source is 0.05 ppm.

The Bengal delta plain is formed by the Ganga–Brahmaputra river system, the 13th largest modern delta in the world. The average annual sediment transport by the Ganga-Brahmaputra river system is about 1,800 tons/km^2 and a suspended load around 540–1,157 million tons per year (with dissolved particulates 173 million tons and suspended solids 1,060 million tons approximately), along with several trace elements channeling toward the Bay of Bengal (Singh 2006; Stephane and Charlet 2007). Arsenic in groundwater is often associated with geologic sources, but in some cases, anthropogenic inputs can be extremely important (Dey et al. 2005; Bhattacharya et al. 2007). However, a specific source of arsenic is yet to be identified in the region, but researchers envisaged few potential minor sources of arsenic, which are located in the Ganges catchment areas including the Himalayas and peninsular India (Acharyya and Shah 2007). The arsenic problem in groundwater in West Bengal, India, was first recognized in the early 1980s (Garai et al. 1984), and the health effects are now reasonably well documented. In fact, more than 50 million people are undoubtedly at risk in the Bengal delta plain (Sharma et al. 2006; Das et al. 2009). More recently, the scale of the problem in other states adjoining the West Bengal with a similar hydrogeologic pattern like Assam, Tripura, Manipur, Arunachal Pradesh, Nagaland, Bihar, Jharkhand, and Uttar Pradesh has also been reported (Mukherjee et al. 2006; Nickson et al. 2007). In northeastern India, the presence of arsenic has been identified in 21 districts out of 24 districts of Assam and in three districts in Tripura, six in Arunachal Pradesh, one in Manipur, and two in Nagaland (Singh 2004; Mukherjee et al. 2006; Das et al. 2009). However, the problem of arsenic in groundwater and related health hazards in Assam is yet to get enough systematic attention due to its recent emergence having few review reports (Singh 2004; SOES 2004; Mukherjee et al. 2006; Nickson et al. 2007; Chetia et al. 2008).

Along with arsenic, the problem of iron in water is another major issue in the region (Singh 2004; Ground Water Information Booklet 2008). Chronic and excess iron consumption is toxic for the health, which might cause genetic disorders like hemochromatosis. This redox flexible element produces oxygen free radicals that are toxic to the cells. Moreover, iron hydroxides in water are supposed to help in the generation of arsenic species (Bhattacharjee et al. 2005). Recently, the problem of manganese has also come up in some parts of Assam (unpublished observation). Numerous pathologic conditions could occur as a consequence of excess persistent intake of manganese including behavioral changes and other nervous system disorders (Elster et al. 1988; ATSDR 2008).

Nowadays in Assam, family- and/or community-based groundwater tubewells have become more popular as a source of potable water instead of surface water for household utilization to do away with the difficulty of water scarcity in the dry period and waterlogging and inundation problems in the rainy season so as to avoid diarrhea and other waterborne diseases. The objective of the present detailed survey-based study was to determine the contamination of heavy metals like arsenic, iron, and manganese and their depth-wise distribution in the tubewell water of Golaghat district (26.0° N to 27.1° N and 93.0° E to 94.18° E) located in the state of Assam. Moreover, the concentrations of other key water quality parameters, namely, sodium (Na), potassium (K), calcium (Ca), magnesium (Mg), pH, total hardness (TH), and sulfate (SO_4^{2-}) were also been examined. Thus, the current article would help to fling light on the water quality especially relating to toxic metals in one of the most important areas of northeast India.

Experimental

Study site

Samples were collected from different regions of Golaghat district of Assam, India. The total geographical area of the district is 3,502 km^2, having tropical, hot, and humid climates with an average annual rainfall of around 2,012 mm/year. Physiographically, the district has a general elevation of around 100 m above mean sea level with a rise at the southern part, where it merges with the hills of the Nagaland as well as Karbi-Anglong district of Assam. Predominant geologic formations include Quaternary Formation (comprises younger and older alluvial deposits consisting of different grades of sand, pebbles, cobbles, gravel, and clay in the area) followed by Archaean Age deposit with rock types like granite, granite gneiss, and quartzite (Ground Water Information Booklet 2008). Older alluvial deposits occur mainly toward southern parts. The hard crystalline of the Archaean Age covers the extreme southern boundary of the district merging with Karbi-Anglong district (Ground Water Information Booklet 2008). Two prominent soil types (light gray to dark gray and deep reddish) are seen in the district with low nitrogen, low phosphate, and medium to high potash contents. Acidic characters of the soil are representative of the soil cover found in the hills. In the plain areas, the other type of the soil covers is found to be feebly alkaline. About 40% of the total district area is covered by forest, 18% by uncultivable land, 2% by fallow land, and 40% by total cropped area with paddy, the principal crop grown in the district (Ground Water Information Booklet 2008). The district is having a unique water regimen with rivers Dhansiri and Kakodonga traversing along the region from south to north direction. The mighty river Brahmaputra flows along the north part of the district. River Kakodonga marks the eastern border of Golaghat and adjoining district Jorhat. Dhansiri, originating from "Laisang peak" of the adjoining state Nagaland, flows northward along the midwestern side of the Golaghat district for a distance of 352 km before joining the Brahmaputra and has a catchment area of 1,220 km^2. Doyang, Nambor, Doigrung, and Kalioni are the four rivulets of Dhansiri. Doyang originates from "Maw" of Nagaland and joins with Dhansiri at the south of Golaghat town, the district headquarters. The other rivulets originate from the adjacent district Karbi-Anglong and flow in a west-to-northeast direction. Hydrogeologically, the district is proved to be very potential, harboring approximately 3–9 prolific aquifer systems. Depth-to-water level measurements in major parts of the district varies from 2 to 7 m. Groundwater occurs under water table to confined conditions as aquifers that tend to increase toward the north and in the southeastern parts (Ground Water Information Booklet 2008).

Tubewell water samples were collected from Golaghat district; the area of the collection appeared under different blocks, namely, Dergaon (Golaghat North), Kathalguri (Golaghat Central), Podumoni (Golaghat East), Kakodonga, Morangi, and Gamariguri within the said district (Fig. 1).

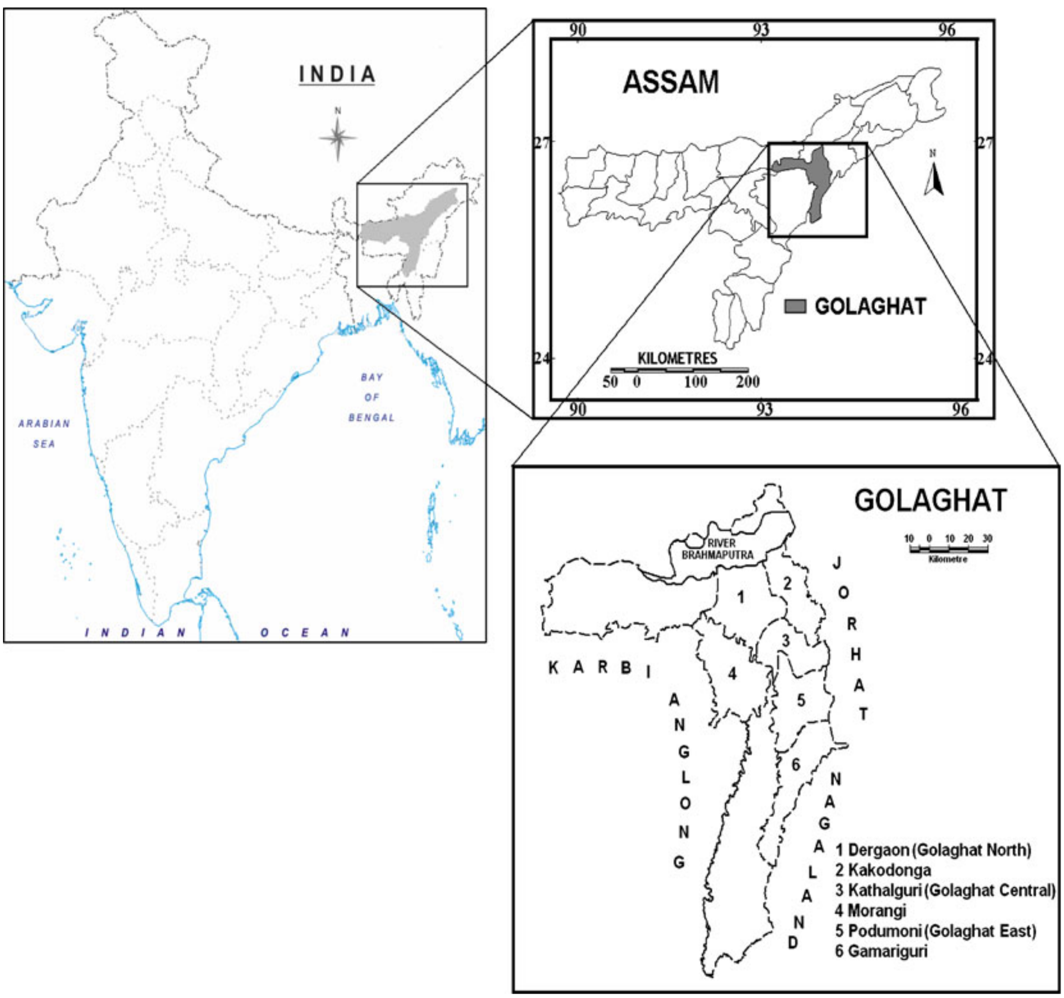


Fig. 1 Study sites showing six blocks of Golaghat district, Assam

Sampling methodology

A total of 222 water samples were collected from the study area during the presummer season (February–March 2008). Maximum samples were collected from Podumoni ($n=72$), followed by Kathalguri ($n=64$), Dergaon ($n=33$), Kakodonga ($n=22$), Morangi ($n=19$), and Gamariguri ($n=12$). The sources of the water samples included shallow hand tubewells (∼60 ± 10 ft deep) and deep tubewells like Tara pump (∼120 ± 10 ft deep) and Mark tubewells (∼180 ± 10 ft deep). Majority of the tubewells (75%) for the collection of samples were for community use. There were 52, 87, and 83 water samples collected from tubewells of different depths, viz., ∼60 ± 10, ∼120 ± 10, and ∼180 ± 10 ft, respectively. In Gamariguri, all samples were collected from deep tubewells having 120- and/or 180-ft depth. It can also be noted that, the Tara pump and the Mark tubewells were mainly installed where water table is lower and through public funds and were utilized by the locality, as the cost of installation of these deep tubewells were too high to bear by a common single individual in the villages. Tubewells were operated for 5 min before collection to wash out the stagnant water inside the tube and to get fresh groundwater. The water samples were collected in clean 1-l polypropylene bottles.

Sample analysis

The collected samples were analyzed for the parameters, viz., As, Fe, Mn, Ca, Na, K, Mg, pH, TH, and SO_4^{2-}. pH of the samples was measured at the site of collection by using the Pocket pH meter (Merck, India), previously standardized by pH 4, 7, and 9 standard buffer solutions (procured from Merck, India). After determination of pH, 1:1 HNO_3 solution was added to each of the water samples collected (to make pH < 2.0), and the samples were carried to the laboratory for further analysis. The analyses of TH, SO_4^{2-}, Na, K, Ca, and Mg were determined following standard methods (Eatson et al. 2005). The instruments were calibrated and standardized before carrying out the analysis. Na and K in the water samples were determined by a flame photometer (Systronics, Germany), whereas TH, Ca, and Mg were determined by ethylenediaminetetraacetic acid titrametric methods. SO_4^{2-} content present in the water samples were analyzed turbidimetrically at 450 nm using a UV–spectrophotometer (SPECORD 40, Analytic Jena, Germany). The concentrations of the heavy metals, namely, Fe and Mn, were determined using atomic absorption spectrometry (AAS; model Perkin Elmer 200, USA) at their respective wavelength and slit width. Hydride Generation-Atomic Absorption Spectrometry (HG-AAS; Perkin Elmer 200, USA) was used for analysis of As in water samples (detection limit 0.02 μg/L). All the reagents were of analytical grade and were purchased from Merck, India, which were prepared freshly at the time of analysis. Standards were procured from Perkin Elmer, USA. A blank was analyzed between element-specific standard readings to verify baseline stability of the instrument. After a batch of ten samples was analyzed, standard solution was additionally analyzed to confirm the calibration of the instrument. For better sensitivity, As^{5+} was prereduced to As^{3+} before analysis. Prereduction was carried out following the user guide of AAS (Eatson et al. 2005). Briefly, a mixture of 5 ml of potassium iodide and ascorbic acid solution, 10 ml of 5-mol/L HCl solution, and 10 ml of water sample was added in a 50-ml volumetric flask. The volume was made up to the mark of the flask with 0.15-mol/L HCl solution. Time given for prereduction was 30 min. Ten milliliters of the prereduced water sample was analyzed using AAS with MHS-15 (Mercury Hydride System) at 193.7 analytical wavelengths and 0.7-nm slit width. Radiation source was an electrode less discharge lamp for arsenic with 20-s prereaction purge time and 10-s postreaction purge time. Argon gas and sodium borohydrate were used for hydride generation. Oxyacetylene flame was used for determination of heavy metals.

Statistical analysis

The data were subjected to Pearson correlation and cluster analysis using Origin (Ver. 6.1), SPSS (Ver. 14.0), and Statistica (Release 7). In this study, cluster analysis was done by applying average linkage (between groups) and Pearson correlation method (hierarchical clustering), which was used as the measure of similarity. Before analyzing for clusters, the data were subjected to Pearson correlation between different water quality parameters of the study site.

Results

The present paper deals with the study of tubewell water samples that were collected from six different blocks of Golaghat district, Assam (Fig. 1). The details of the study site and the determination of water quality parameters were presented in "Experimental" of this paper. Table 1 represented the concentration of different elements like As, Fe, Mn, and Ca in the groundwater samples collected from tubewells of the six blocks, namely, Dergaon, Kathalguri, Podumoni, Kakodonga, Morangi, and Gamariguri of Golaghat district. Concentration of As in the groundwater samples, collected from tubewells, differed in different blocks: Dergaon (Trace to 0.087 ppm), Kathalguri (Trace to 0.089 ppm), Podumoni (Trace to 0.123 ppm), Kakodonga (Trace to 0.06 ppm), Morangi (Trace to 0.07 ppm), and Gamariguri (0.074–0.128 ppm; Table 1). The most badly affected area was Gamariguri block, where 100% of the samples ($n = 12$), collected from deep tubewell waters, had an As concentration above the WHO/BIS guideline value of As (0.01 ppm;

Table 1 Concentrations of As, Fe, Mn, and Ca in tubewell waters of the study site

Blocks	Parameters	As (ppm)	Fe (ppm)	Mn (ppm)	Ca (ppm)
	WHO/BIS DL	0.01	0.3	0.1	75
	WHO/BIS PL	No relaxation[a]	1.0	0.3	200
Dergaon	Range (ppm)	Tr–0.087	0.43–5.8	Tr–2.6	3.2–82.4
	Mean ± SD	0.019 ± 0.02	2.7 ± 1.9	0.51 ± 0.6	19.7 ± 17.3
	Below DL (%)	57.6	0	24.2	96.97
	Within WHO/BIS limit (%)	0	21.2	33.4	0
	Above PL (%)	42.4	78.8	42.4	3.03
Kathalguri	Range	Tr–0.089	0.28–5.9	Tr–0.61	0–60.8
	Mean ± SD	0.03 ± 0.02	3.8 ± 1.6	0.13 ± 0.15	20.1 ± 9.5
	Below DL (%)	28.1	1.6	56.3	100
	Within WHO/BIS limit (%)	0	14	28.1	0
	Above PL (%)	71.9	84.4	15.6	0
Podumoni	Range	Tr–0.123	0.34–5.9	Tr–0.98	3.2–56.8
	Mean ± SD	0.06–0.04	3.21–1.5	0.17–0.16	23.1–10.4
	Below DL (%)	12.5	0	47.2	100
	Within WHO/BIS limit (%)	0	13.9	33.4	0
	Above PL (%)	87.5	86.1	19.4	0
Kakodonga	Range	Tr–0.06	0.1–3.5	0.006–1.9	4–44.8
	Mean ± SD	0.02–0.02	1.18–0.96	0.36–0.5	19.2–11.8
	Below DL (%)	63.6	13.6	45.5	100
	Within WHO/BIS limit (%)	0	40.9	31.8	0
	Above PL (%)	36.4	45.5	22.7	0
Morangi	Range	Tr–0.07	0.32–3.8	0.012–0.98	4–36
	Mean ± SD	0.016–0.02	1.4–0.9	0.26–0.23	14.03–8.1
	Below DL (%)	57.9	0	31.6	100
	Within WHO/BIS limit (%)	0	36.8	47.3	0
	Above PL (%)	42.1	63.2	21.1	0
Gamariguri	Range	0.074–0.128	3.5–5.9	0.02–2.5	9.6–30.4
	Mean ± SD	0.10–0.02	4.7–0.8	0.92–0.9	19.6–7.4
	Below DL (%)	0	0	16.7	100
	Within WHO/BIS limit (%)	0	0	33.3	0
	Above PL (%)	100	100	50	0

DL desirable limit, *PL* permissible limit, *Tr* trace

[a]Maximum permissible limit in the absence of an alternative source is 0.05 ppm (BIS 1991)

WHO 1993; BIS 1991) followed by Podumoni (87.5%), Kathalguri (71.9%), Dergaon (42.4%), Morangi (42.1%), and Kakodonga (36.4%). Maximum As content recorded was 0.128 ppm in a water sample collected from Gamariguri block. It was also noted that all samples collected from the Gamariguri block were from deep tubewells. Again, the analysis showed that 76.4% of the groundwater samples of the study area had Fe content higher than the permissible limit (1.0 ppm) provided by the WHO/BIS (WHO 1993; BIS 1991); Gamariguri (100%), Podumoni, (86.1%), Kathalguri (84.4%), Dergaon (78.8%), Morangi (63.2%), and Kakodonga (45.5%) were worse affected. The wide variation of Fe content in the collected samples was represented in Table 1 with lowest concentration observed in a sample collected from Kakodonga (0.1 ppm) and the highest, 5.9 ppm, in three samples of Kathalguri, Podumoni, and Gamariguri (one in each block). The study also revealed that 28.5% of total groundwater samples collected from different tubewells had Mn concentration above the permissible limit (0.3 ppm) of the WHO/BIS (WHO 1993; BIS 1991) standards. The highly contaminated area was the Gamariguri block, where 50% groundwater samples had Mn content above the permissible limit, followed by Dergaon (42.4%), Kakodonga (22.7%), Morangi (21.1%), Podumoni (19.4%), and Kathalguri (15.6%). The

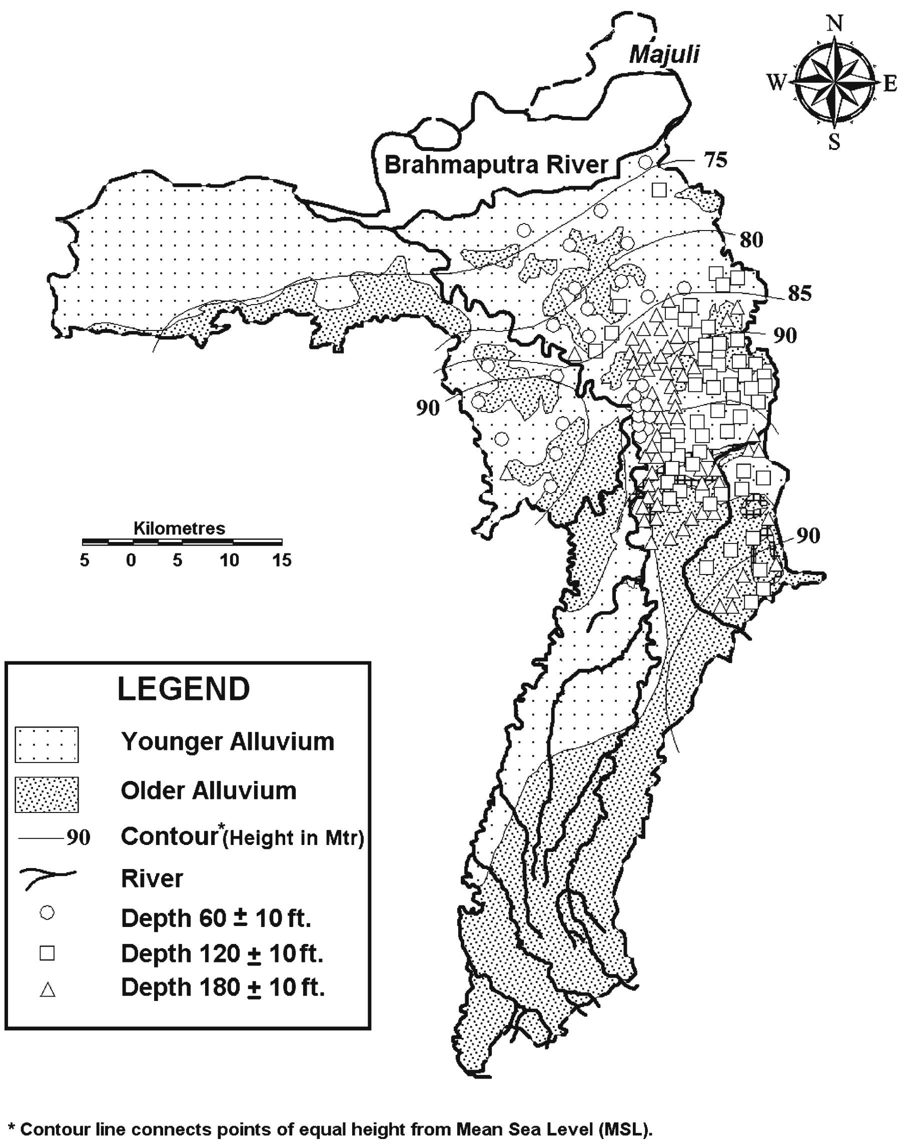
Table 2 Concentration of Na, K, Mg, pH, TH, and SO_4^{2-} in tubewell waters of the study site

Blocks	Parameters	Na (ppm)	K (ppm)	Mg (ppm)	pH	TH (ppm)	SO_4^{2-} (ppm)
	WHO/BIS DL	200.0	n.a.	30	6.5	300	200
	WHO/BIS PL	No relaxation	n.a.	100	8.5	600	400
Dergaon	Range	4.5–28.7	1.26–4	5.76–55.4	5.9–7.7	34.9–310.4	12.5–62.0
	Mean ± SD	12.6 ± 5.7	8.7 ± 8.6	25.8 ± 11.6	6.9 ± 0.4	126 ± 61.2	28.4 ± 10.1
	Below DL (%)	100	–	69.7	12.12	96.97	100
	Within WHO/BIS limit (%)	0	–	30.3	87.9	0	0
	Above PL (%)	0	–	0	0	3.03	0
Kathalguri	Range	10.3–68.1	0.75–15.3	Tr–32.02	6.2–8.1	38.8–176	0.34–86.2
	Mean ± SD	21.3 ± 9.6	2.2 ± 1.9	13.3 ± 8.7	7.1 ± 0.35	106.7 ± 30.1	15.9 ± 21.5
	Below DL (%)	100	–	96.9	4.7	100	100
	Within WHO/BIS limit (%)	0	–	3.12	95.32	0	0
	Above PL (%)	0	–	0	0	0	0
Podumoni	Range	0.14–204.4	0.5–9.4	0.25–52.8	6.2–7.6	7.8–250	0.38–108.8
	Mean ± SD	59.3–42.5	1.58–1.1	23.9–8.5	7.1–0.3	121.0–42.6	45.1–26.1
	Below DL (%)	100	–	79.2	41.6	100	100
	Within WHO/BIS limit (%)	0	–	20.8	58.3	0	0
	Above PL (%)	0	–	0	0	0	0
Kakodonga	Range	6.9–29.5	1–5.92	4.5–37.8	6.8–7.5	23.3–197.8	12.35–65.3
	Mean ± SD	17.3–7.1	2.6–1.4	20.8–10.8	7.05–0.2	104.1–55.3	32.8–18.1
	Below DL (%)	100	–	95	0	100	100
	Within WHO/BIS limit (%)	0	–	5	100	0	0
	Above PL (%)	0	–	0	0	0	0
Morangi	Range	5.9–67.3	1.44–49.1	0.68–25.7	6.8–7.5	23.3–131.9	2.3–63.2
	Mean ± SD	20.0–13.8	8.27–10.6	12.06–6.5	7.1–0.2	64.7–33.05	23.9–18.04
	Below DL (%)	100	–	100	0	100	100
	Within WHO/BIS limit (%)	0	–	0	100	0	0
	Above PL (%)	0	–	0	0	0	0
Gamariguri	Range	4.8–175.7	1–1.66	4.9–27.1	6.9–7.1	50.4–139.7	15.3–98.3
	Mean ± SD	62.2–58.1	1.29–0.3	16.5–6.6	7.1–0.1	89.8–31.0	53.3–22.8
	Below DL (%)	100	–	100	0	100	100
	Within WHO/BIS limit (%)	0	–	0	100	0	0
	Above PL (%)	0	–	0	0	0	0

DL desirable limit, *PL* permissible limit, *Tr* Trace highest recorded value of Mn was 2.6 ppm in a sample from the Dergaon block. The Mn content in Dergaon, Kathalguri, Podumoni, Kakodonga, Morangi, and Gamariguri blocks varied from Trace to 2.6 ppm, Trace to 0.61 ppm, Trace to 0.98 ppm, 0.006 to 1.9 ppm, 0.012 to 0.98 ppm, and 0.02 to 2.5 ppm, respectively (Table 1). It was observed from the experiment that the pH of the water samples collected in the Dergaon (5.9 to 7.7), Kathalguri (6.2 to 8.1), Podumoni (6.2 to 7.6), Kakodonga (6.8 to 7.5), Morangi (6.8 to 7.5), and Gamariguri (6.9 to 7.1) blocks were varied considerably (Table 2). The concentrations of the other parameters, viz., Ca, Na, K, Mg, TH, and SO_4^{2-} (Tables 1, 2) in all the samples collected from the study area were within the safe limit provided by the WHO/BIS (WHO 1993; BIS 1991). However, the pH of 90.3% of the samples was found within the drinking water standard (6.5–8.5) of the WHO/BIS. The maximum value of the pH was recorded as 8.1 in a sample from the Kathalguri block.

Figure 2 represented the alluvial pattern of Golaghat district, showing younger and older alluviums, including the sites having higher concentration of As. However, the study site also revealed

Fig. 2 Showing the alluvial pattern of Golaghat district and the sampling sites and depth of tubewells where the concentrations of As and Fe are higher than the drinking water guideline value of the WHO/BIS



that the As and Fe contaminations of tubewell waters were mostly found on the younger alluvium itself (Fig. 3). It was also noticed that the depth-wise distribution and contamination level of As above the permissible limit in tubewell waters followed an interesting pattern (Fig. 4). Seventy-eight percent of the total 83 samples collected from the tubewells, with depth 180 ± 10 ft, showed an As concentration that was higher than the limit value of the WHO/BIS. Similarly, 64% ($n = 87$) and 58% ($n = 52$) of the samples, collected from tubewells of depths 120 ± 10 and 60 ± 10 ft, respectively, had an As concentration above the WHO limit and BIS limit (Table 3). However, higher than the permissible limit (≤1 ppm for drinking water) for Fe was found in case of 83%, each for the samples collected from 180 ± 10 and 120 ± 10 ft, respectively, and 65% for samples collected from depth 60 ± 10 ft, showing high level of Fe contamination in the groundwater tables (Table 4). However, interestingly, approximately 25% of the samples collected from tubewells of

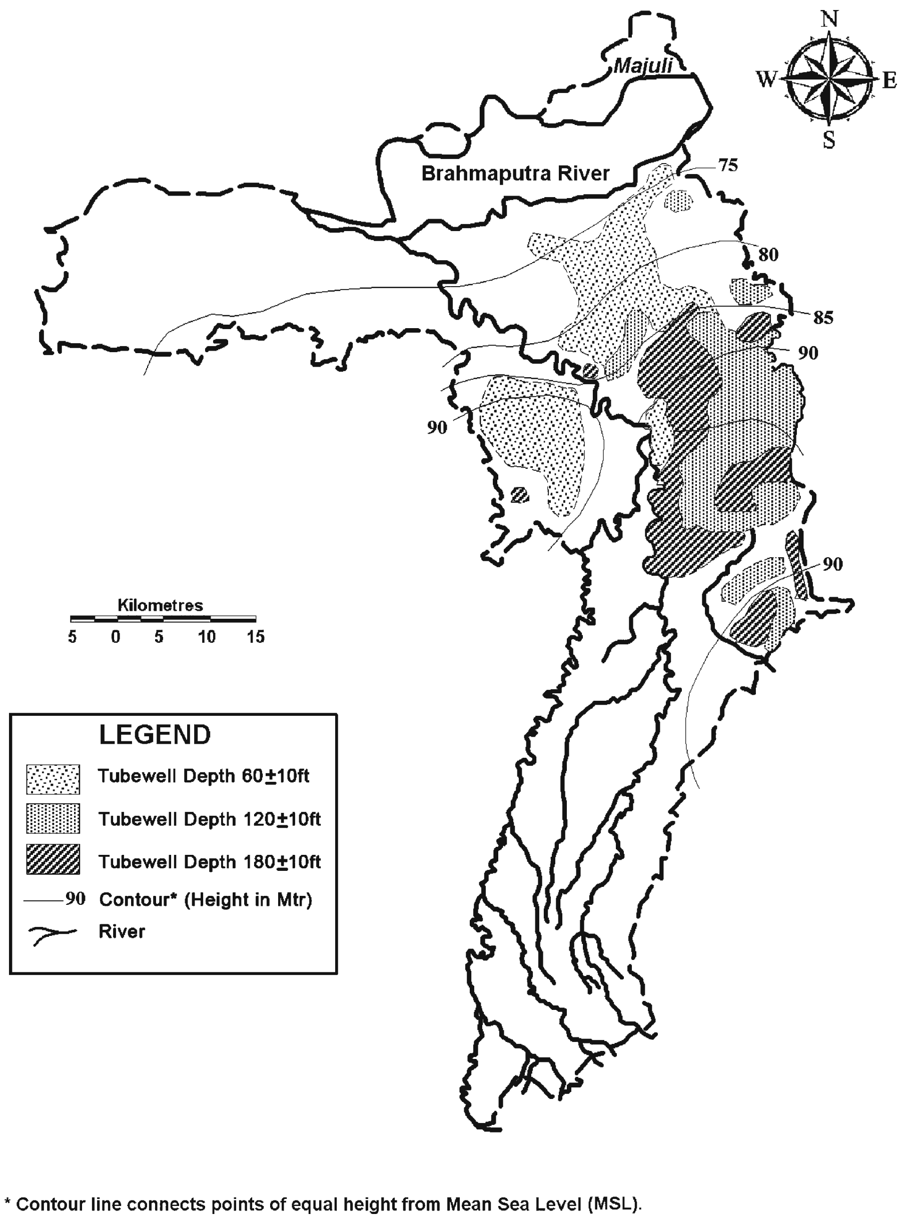


Fig. 3 Showing As and Fe contaminated tubewell sites at different depth of the study area

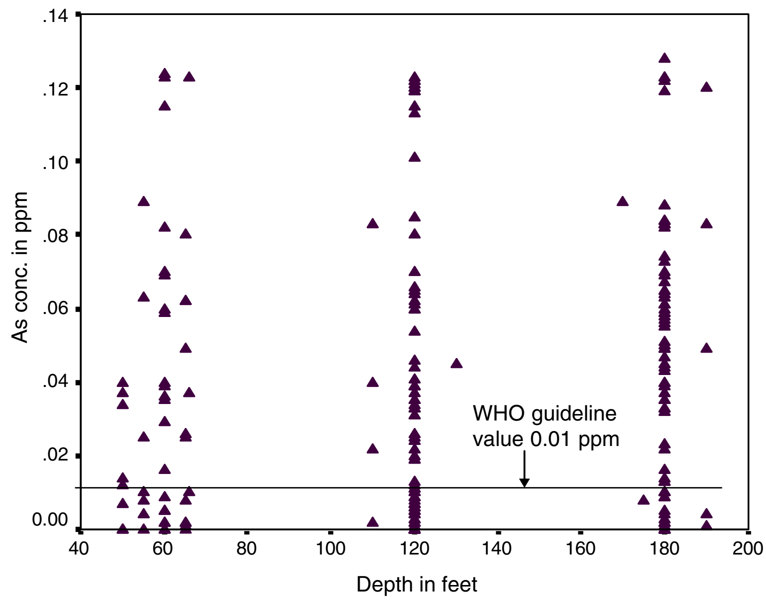


Fig. 4 Graphical representation of depth-wise As distribution and concentration in tubewell waters of the study site

different depths showed an Mn concentration higher than the prescribed value by the WHO/BIS (Table 4).

Statistical analysis using Pearson correlation showed that the parameters in the tubewell water samples collected from Golaghat were weakly and moderately correlated to each other at $p < 0.01$ and $p < 0.05$ levels. A significant positive correlation was found to exist between As and Fe (0.697), Mn (0.212), Na (0.488), Ca (0.212), Mg (0.197), and SO_4^{2-} (0.498) at $p < 0.01$ (Table 5). Correlations between Fe and Mn (0.213), TH (0.216), Na (0.329), and Ca (0.254) at $p < 0.01$ were also observed in the collected water samples of the study region. Similarly, some correlations were also observed (Table 5) between Mn and Ca (0.218), Mg (0.168), SO_4^{2-} (0.216), pH and Ca (0.236), Na and SO_4^{2-} (0.302), and Mg and SO_4^{2-} (0.309) at $p < 0.01$ level. Again, a strong positive correlation was found between TH and Ca (0.755) and Mg (0.793) at $p < 0.01$. From Table 5. it was seen that Ca was correlated significantly with Mg, having a correlation coefficient of 0.550 at $p < 0.01$. In this study, cluster analysis was employed to detect similarity and dissimilarity groups between the water quality parameters of the collected groundwater samples from the six blocks of Golaghat district. The cluster analy-

Table 3 Depth-wise As distribution in tubewell waters of different blocks of Golaghat

Blocks	60 ± 10 ft			120 ± 10 ft			180 ± 10 ft		
	<0.01 ppm	0.01–0.05 ppm	>0.05 ppm	<0.01 ppm	0.01–0.05 ppm	>0.05 ppm	<0.01 ppm	0.01–0.05 ppm	>0.05 ppm
Kakodonga	7 (10)	3 (10)	0 (10)	5 (8)	2 (8)	1 (8)	2 (4)	2 (4)	0 (4)
Morangi	4 (10)	5 (10)	1 (10)	3 (4)	1 (4)	0 (4)	4 (5)	1 (5)	0 (5)
Dergaon	6 (16)	4 (16)	6 (16)	10 (13)	2 (13)	1 (13)	3 (4)	1 (4)	0 (4)
Kathalguri	3 (4)	0 (4)	1 (4)	10 (32)	21 (32)	1 (32)	5 (28)	16 (28)	7 (28)
Podumoni	2 (12)	4 (12)	6 (12)	3 (24)	3 (24)	18 (24)	4 (36)	8 (36)	24 (36)
Gamariguri	0	0	0	0 (6)	0 (6)	6 (6)	0 (6)	0 (6)	6 (6)
Total samples	22 (52)	16 (52)	14 (52)	31 (87)	29 (87)	27 (87)	18 (83)	28 (83)	37 (83)
Percentage (%)	42.3%	30.8%	26.9%	35.6%	33.3%	31.1%	21.7%	33.7%	44.6%

The number within brackets indicates the size of the samples collected depth-wise from the respective block under examination

Table 4 Depth-wise distribution of number of samples above the permissible limit (WHO/BIS limit) for total Fe and Mn, in tubewell waters of the study area

Blocks	60 ± 10 ft		120 ± 10 ft		180 ± 10 ft	
	Fe (>1 ppm)	Mn (>0.3 ppm)	Fe (>1 ppm)	Mn (>0.3 ppm)	Fe (>1 ppm)	Mn (>0.3 ppm)
Kakodonga	3 (10)	3 (10)	5 (8)	1 (8)	2 (4)	1 (4)
Morangi	6 (10)	1 (10)	2 (4)	1 (4)	4 (5)	2 (5)
Dergaon	13 (16)	6 (16)	9 (13)	3 (13)	4 (4)	1 (4)
Kathalguri	2 (4)	1 (4)	29 (32)	4 (32)	22 (28)	5 (28)
Podumoni	10 (12)	3 (12)	21 (24)	6 (24)	31 (36)	5 (36)
Gamariguri	0	0	6 (6)	6 (6)	6 (6)	6 (6)
No. of samples above WHO/BIS limit	34 (52)	14 (52)	72 (87)	21 (87)	69 (83)	20 (83)
Percentage of samples above WHO/BIS limit	65.4%	26.9%	82.7%	24.1%	83.1%	24.1%

The number within brackets indicates the size of the samples collected depth-wise from the respective block under examination

Table 5 Pearson correlation between different water quality parameters of the study site

	As	Fe	Mn	pH	TH	Na	K	Ca	Mg	SO_4^{2-}
As	1									
Fe	0.697[a]	1								
Mn	0.212[a]	0.213[a]	1							
pH	0.114	0.063	−0.057	1						
TH	0.152[b]	0.216[a]	0.157[b]	0.141[b]	1					
Na	0.488[a]	0.329[a]	−0.130	0.112	−0.006	1				
K	−0.206[a]	−0.172[b]	0.038	−0.101	0.072	−0.175[a]	1			
Ca	0.212[a]	0.254[a]	0.218[a]	0.236[a]	0.755[a]	0.014	−0.090	1		
Mg	0.197[a]	0.074	0.168[b]	0.022	0.793[a]	0.065	0.158[b]	0.550[a]	1	
SO_4^{2-}	0.498[a]	0.159[b]	0.216[a]	0.007	0.096	0.302[a]	−0.090	0.117	0.309[a]	1

[a]Correlation is significant at the 0.01 level (two tailed)
[b]Correlation is significant at the 0.05 level (two tailed)

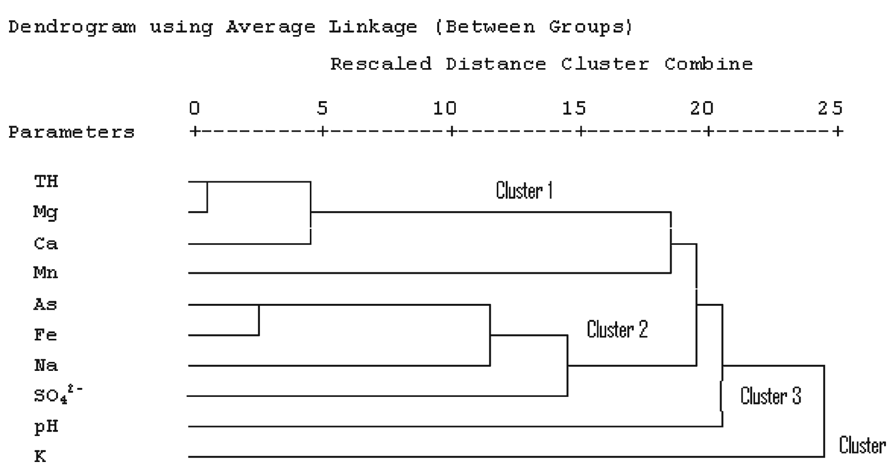


Fig. 5 Dendrogram showing the clusters of different water quality parameters of the study site

sis was done by using average linkage (between groups) and Pearson correlation methods. The dendrogram in Fig. 5 showed four distinct clusters of water quality parameters of the study area. Cluster 1 includes the water quality parameters TH, Mg, Ca, and Mn. Cluster 2 included As, Fe, Na, and SO_4^{2-}. These two clusters (clusters 1 and 2) were connected with cluster 3 comprising pH, which is again connected with cluster 4 having only one parameter, K.

Discussion

The present study indicated the distribution of different water quality parameters including heavy metals like As, Fe, and Mn of the tubewells in the six blocks of Golaghat district of Assam, India. It was revealed that the residents of the area had been chronically exposed to high levels (above the permissible limit of the WHO/BIS) of As (67% of the total 222 samples collected) and Fe (77% of the total 222 samples collected), in comparison with the highly As-exposed populations of the adjoining regions like West Bengal, India (Mukherjee et al. 2006; Nickson et al. 2007) and Bangladesh (Bhattacharya et al. 2007; Halem et al. 2009). Again, the study also revealed that around 35.1% of the water sources of tubewells have an As contamination of more than the 0.05 ppm, which is higher (11.2%) than the previous report by Nickson et al. (2007). It was apparent from the study for As in the region that groundwater adjacent to the foothills (Gamariguri block) is highly As-contaminated as mentioned by another author (Singh 2004). This area is mainly within the quaternary alluvial basin (Ground Water Information Booklet 2008) bounded by the Himalayan Mountains. However, the Kakodonga block, bordering the river Kakodonga, was less contaminated with As with 63.6% and Fe with 45.5% of the water samples, within the safe limit for drinking purposes (Table 1). The probable reason for As contamination might be heavy deposition of sediments through leaching from surrounding mountains (Singh 2004). In addition, the present study revealed that the quaternary alluvium deposits especially the younger regions were more As-contaminated, in comparison to the older one (Figs. 2, 3). Our findings also corresponded the findings of earlier workers (McArthur et al. 2004; Mukherjee et al. 2006) who had reported that the As-contaminated zones in the GPMB river basin are mostly lying within a younger alluvium plain.

Mobilization of As in tubewell waters in the Gangetic delta of West Bangal, India and Bangladesh had already been proposed by several workers (Smedley and Kinniburgh 2002; Rmalli et al. 2005; McArthur et al. 2004; Bhattacharya et al. 2006). Regarding sedimentology, the north-eastern part of India, especially Assam region, is related to that of the Bangladesh plains (Enmark and Nordborg 2007). Therefore, the adjoining district Golaghat, the site of our interest, might be expected to follow the comparable way in respect to the mobilization of As in groundwater. Workers like Smedley and Kinniburgh (2002), McArthur et al. (2004), and Bhattacharya et al. (2006) proposed the importance of reductive dissolution of metal (iron) oxide/hydroxide and subsequent release of the adsorbed As, in the process of its mobilization. The degradation of organic matter present in the aquifers might also be involved in the reductive dissolution of the iron hydroxide, causing desorption of any adsorbed As (Parkhurst 1995). Moreover, Fe hydroxides, carrying a surface charge, could adsorb the electrically charged As ions that appeared to be a pH-dependent process and related to the net charge of the adsorbing surface (Parkhurst 1995). Generation of As oxyanionic species through oxidative dissolution of the As-bearing pyrite minerals ($FeAsS + 3.5O_2 + 4H_2O \rightarrow Fe(OH)_3 + H_3AsO_4 + 2H^+ + SO_4^{2-}$) could be the another basis of contamination of the groundwater tables (Acharyya et al. 1999; Nickson et al. 2000). In this study, higher concentrations of As and Fe (Table 1) in the tubewell waters of Golaghat district showed a significant correlation between the two elements (0.697 at $p < 0.01$; Table 5). The phenomenon of the mobilization of As might be due to the reductive dissolution of As–Fe-bearing minerals in the sediments reduced by oxygen-deficient groundwater (Singh 2004). Overwithdrawal of groundwater in the study region for agriculture practices and household uses might be another strong reason for the As mobilization (Bhattacharjee et al.

2005) in aquifers in Golaghat district. However, the presence of SO_4^{2-} in tubewell waters in the region having a significant correlation (0.498 at $p < 0.01$; Table 4) with As could also be one of the probable sources of As mobilization through which As might possibly come to the water environment by the oxidation of the sulfide ores of As (Acharyya et al. 1999). Again, unhygienic soap use in tubewells area facilitated microorganisms to release arsenic-involving bioelectrochemical reactions (Dey et al. 2005). A significant correlation between As with Fe and Mn (Table 5) indicated that the tubewell water of the study area might have some common natural sources of pollution through which mobilization of As, Fe, and Mn was taking place.

The region has the potential resources of water in so far as groundwater is concerned (1,056.26 million cubic meters for the district as mentioned in Ground Water Information Booklet 2008), the area belonging to northern part of the district like the Dergaon, Kakodonga, and Morangi blocks. The groundwater level in these three blocks was found to be closer as mainly shallow tubewells were found during the collection of the samples. Moreover, availability and utilization of surface water for household purposes might be the basis of the fewer number of tubewells in the region.

The incidence of As contamination (>0.01 ppm) was noted greater in the southern parts of the districts compared to the northern part. Again, 78% of the water collected from the tubewells having a depth of 180 ± 10 ft was found to be contaminated by As, followed by 65% of the water from 120 ± 10 ft. Potential aquifers pertained to the Quaternary Formation have unique subsurface geology. The cumulative thickness of the aquifer zones of the district has the tendency to increase toward the north and in the southeastern parts; however, the thickness of the upper bedrock reverses considerably with thicker midzones. The movement of groundwater is from south to north, and the trend of the water level shows a gradual rising toward the north. According to the hydrogeologic information of the Central Ground Water Board, India, there are possibly 3–9 prolific aquifer systems that exist in the district (Ground Water Information Booklet 2008), and these aquifers might be the sources of As contamination of the tubewells in the study region. The layers of aquifers possibly arranged in a horizontal pattern in relation to the slope of the district usually get recharged from distant sources. Little mixing occurs across these flow-paths, and therefore, solute concentrations within an aquifer were also layered, with each deeper layer representing the biogeochemical outcome of water inputs from more remote sources (Harvey 2008). It had been elaborated in a recent study (Polizzotto et al. 2008) that pond sediments were also an important source of As mobilization in groundwater. The steady settling and decomposition of organic matter at the bottom of the tropical ponds, like the area of interest, in an anaerobic condition ensuing microbe mediated Fe (III) and As (V) reductions (McArthur et al. 2004; Polizzotto et al. 2005, 2008). Furthermore, water passing through pond sediments could also contain organic carbon that, on decomposition, might help to liberate arsenic from deeper sediments, adding more to the contamination (Harvey 2008).

Conclusion

For almost two decades, research on As has gained a considerable momentum as a response to the detrimental health effects of the element. The recognition of the scale of As enrichments in groundwater in West Bengal, India and Bangladesh and elsewhere has opened up a serious concern in the scientific community. Billions of people use to drink waters from aquifers daily; however, new reports are coming from different areas regarding As contamination. Several factors are involved in the ever expanding impure water tables throughout the world, involving new aquifers that are yet to be recognized. This paper represented the detailed groundwater quality in the different blocks of Golaghat district. Around 35% of the water sources of the tubewells were found having an As contamination of more than the 0.05 ppm and 67% above 0.01 ppm. Again, a high percentage of Fe contamination (77%) in the tubewell waters of the region indicates the severity of the heavy metal pollution in the region.

Therefore, the primary concern to counter the problem of groundwater contamination especially with a high-priority toxic substance like As in a newly reported region is an early survey-based detection of the pollution and identification of the affected sources to remediate the crisis. The mitigation strategy for the problem in the area might be specific to the location, taking into considerations the geomorphological variations and socioeconomic conditions. Understanding the groundwater movements requires in-depth characterization and routine verification of physical hydrogeology. Moreover, community participation to make the villagers of the affected regions of the district studied understand the signs and symptoms of the chronic As toxic effects is of utmost necessary. Again, cost-effective, user-friendly technologies providing pure water are required to counter the serious health hazards due to consumption of As- and/or Fe-contaminated water. A wholistic approach involving medical practitioners, scientists, and social workers will need to work coherently to find out a solution that can lessen sufferings of the humanity and making a provision for safe drinking water.

Acknowledgements The authors gratefully acknowledge Mr. P. K. Mudoi and Mr. P. K. Sarma Tamuli, Junior Engineer, PHED, Golaghat, Government of Assam and Mr. N. Bora, Project Manager, Arunodoy, Golaghat (NGO) for their kind help and support during sample collection. Two anonymous reviewers are thanked for their excellent comments on the draft manuscript that greatly improved the final version.

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