



Quaternary stratigraphy, sediment characteristics and geochemistry of arsenic-contaminated alluvial aquifers in the Ganges–Brahmaputra floodplain in central Bangladesh

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ABSTRACT

This study focuses on the Quaternary stratigraphy, sediment composition, mineralogy, and geochemistry of arsenic (As)-contaminated alluvial aquifers in the Ganges–Brahmaputra floodplain in the central Bangladesh. Arsenic concentrations in 85 tubewells in Manikganj area, 70 km northwest of Dhaka City, range from 0.25 µg/L to 191 µg/L with a mean concentration of 33 µg/L. Groundwater is mainly Ca–HCO₃ type with high concentrations of dissolved As, Fe, and Mn, but low level of SO₄. The uppermost aquifer occurs between 10 m and 80 m below the surface that has a mean arsenic concentration of 35 µg/L. Deeper aquifer (>100 m depth) has a mean arsenic concentration of 18 µg/L. Sediments in the upper aquifer are mostly gray to dark-gray, whereas sediments in the deep aquifer are mostly yellowing-gray to brown. Quartz, feldspar, mica, hornblende, garnet, kyanite, tourmaline, magnetite, ilmenite are the major minerals in sediments from both aquifers. Biotite and potassium feldspar are dominant in shallow aquifer, although plagioclase feldspar and garnet are abundant in deep aquifer sediments. Sediment composition suggests a mixed provenance with sediment supplies from both orogenic belts and cratons. High arsenic concentrations in sediments are found within the upper 50 m in drilled core samples. Statistical analysis shows that As, Fe, Mn, Ca, and P are strongly correlated in sediments. Concentrations of Cd, Cu, Ni, Zn, and Bi also show strong correlations with arsenic in the Manikganj sediment cores. Authigenic goethite concretions, possibly formed by bacteria, are found in the shallow sediments, which contain arsenic of a concentration as high as 8.8 mg/kg. High arsenic concentrations in aquifers are associated with fine-grained sediments that were derived mostly from the recycled orogens and relatively rapidly deposited mainly by meandering channels during the Early to Middle Holocene rising sea-level conditions.

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1. Introduction

Naturally occurring high concentration of dissolved arsenic (As) in groundwater of alluvial aquifers in Bangladesh has been causing serious health problems in millions of people for the last three decades (Nickson et al., 2000; Smith et al., 2000; BGS and DPHE, 2001; Ahmed et al., 2004; Zheng et al., 2004;

Harvey et al., 2006). In Bangladesh, groundwater is the principal source of drinking and irrigation water supplies. Millions of tubewells were installed in the Ganges–Brahmaputra–Meghna (GBM) delta complex in the last four decades that provided pathogen-free water for domestic and irrigation purposes (Smith et al., 2000; BGS and DPHE, 2001). The major switch from polluted surface water to groundwater in early 1970s helped people avoid waterborne diseases, but detection of elevated dissolved arsenic in groundwater has frightened the people of Bangladesh (Nickson et al., 2000; BGS and DPHE, 2001). In 1993, the Department of Public Health Engineering (DPHE) of Bangladesh first reported the existence of arsenic

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poisoning in groundwater in the southwest of Bangladesh (Dhar et al., 1997; WARPO, 2000; BGS and DPHE, 2001). The National Hydrochemical Survey of Bangladesh (NHS), which was carried out by DPHE and the British Geological Survey (BGS), and Mott MacDonald Ltd. in 1998 and 1999 analyzed 4140 tubewells (including NHS special study areas) for arsenic and other elements (Fig. 1). The survey found that nearly 35 million people were drinking groundwater containing As with a concentration of more than 50 µg/L (Bangladesh standard), and about 57 million people exposed to a concentration exceeding 10 µg/L (World Health Organization standard), mostly extracted from alluvial aquifers located within 10–50 m of the ground surface (BGS and DPHE, 2001).

The occurrence, origin, and mobility of arsenic in groundwater in Bangladesh and many other parts of the world in similar environments are reported (Ravenscroft et al., 2001). The mode of occurrence and mobility of arsenic in sedimentary aquifers are mainly influenced by local geology, geomorphology, hydrogeology, and geochemistry of sediments and water, as well as anthropogenic activities, such as mining and land use (Bhattacharya et al., 1997; BGS and DPHE, 2001; Smedley and Kinniburgh, 2002). In Bangladesh, the occurrence of arsenic and its mobilization is associated with geochemically reducing subsurface environment (Nickson et al., 2000; Zheng et al., 2004). Several recent studies agreed that biogenic reductive dissolution of Fe-oxyhydroxides is the primary release mechanism that puts arsenic into groundwater in Bangladesh alluvial aquifers (Bhattacharya et al., 1997; Nickson et al., 1998; Zheng et al., 2004). A study in the central Bangladesh by Harvey et al. (2002) suggested that arsenic mobilization may also be associated with recent inflow of carbon due to large-scale irrigation pumping, which needs further investigation. Saunders et al. (2005) attempted to link the elevated arsenic occurrences in groundwater with the retreat of continental glaciation at the end of Pleistocene, which led to the rise of sea level during the Early to Middle Holocene, and deposition of alluvium and extensive marsh and peat and finer sediments in Bengal lowlands (Ravenscroft et al., 2001). During the Pleistocene time the mechanical weathering of rocks in source areas (e.g., Himalayas, Indian Shield, and Indo-Burman mountains) was enhanced due to mountain building activities and glaciation. The aquifer sands in the Bengal Basin were largely derived from physical weathering and erosion at a time of extended glaciation in the Himalayas, but the intensity of chemical weathering was limited by low temperatures during erosion (McArthur et al., 2004).

The complex relationships between groundwater arsenic and aquifer mineralogy, geochemistry and sediments characteristics are not adequately addressed. Few studies in Bangladesh and West Bengal, India, found that fine-grained sediments (e.g., clay), and peat in the Holocene deposits contain high arsenic concentrations (e.g., BGS and DPHE, 2001; Pal et al., 2002; Tareq et al., 2003; Horneman et al., 2004; Sengupta et al., 2004). Within the coarse-grained sediment layers, the sand coated by iron-oxyhydroxides, residual magnetite, ilmenite, illite, iron hydroxides-coated grains, biotite, and siderite concretions contain most of the arsenic (Pal et al., 2002; Ahmed et al., 2004). Dissolved arsenic shows a moderate to strong statistical and spatial correlation with iron and phosphorous in groundwaters located under-

neath the Holocene geologic–geomorphic units of Bangladesh (Shamsudduha et al., 2006). Few studies that were performed on heavy mineral assemblages in the arsenic-contaminated aquifers suggested that the arsenic-bearing sediments are primarily derived from the Himalayas and the shield areas. However, the presence of high arsenic in sediments is not associated with primary arsenic-bearing minerals, rather mostly appears to be secondary in origin (Ghosh and De, 1995).

The present study has examined the sediment characteristics, mineralogical composition and geochemistry of the arsenic-affected alluvial aquifers in the Ganges–Brahmaputra floodplains in central Bangladesh. The central objective of this study is to characterize the relationships among elevated concentrations of aqueous arsenic in alluvial aquifers, groundwater and sediment geochemistry with better understanding of aquifer mineralogy, provenance and depositional environments of the Quaternary alluvial sediments in the Bengal Basin.

2. Study area: geology, stratigraphy and hydrogeology

The study area (approximately 40 km²) covers the Manikganj town and its surrounding area, which is located approximately 70 km northwest of Dhaka City, the capital of Bangladesh (Fig. 2a). Geographically, the study area is confined within 23.80° to 23.90° N latitudes, and 89.95° to 90.05° E longitudes, covered mostly by the Ganges–Brahmaputra floodplain deposits. Tectonically, the study area is positioned in the central part of the Bengal Basin, which is the major depocenter of sedimentary flux from the Himalayas and Indo-Burman ranges drained by the Ganges–Brahmaputra–Meghna river system (Goodbred and Kuehl, 2000). The basin is bounded by the Himalayas to the distant north, the Shillong Plateau, a Precambrian massif to the immediate north, the Indo-Burman ranges to the east, the Indian craton to the west, and the Bay of Bengal to the south (Fig. 2; Uddin and Lundberg, 1998a). Surface geology of the Manikganj study area is fairly simple. The entire Manikganj district area is covered with the alluvial silt and clay, alluvial silt, and marsh peat and clay units (Fig. 2b). Manikganj is located in the southern periphery of the Madhupur Tract (a Pleistocene inlier), which is surrounded by the Brahmaputra (Jamuna) depression to the west and Ganges (Padma) depression to the south.

The landscape of the study area is typically fluvial, characterized mainly by active channels, abandoned channels, natural levees, backswamps, and floodplains. Distribution of the geomorphic features throughout the study area suggests that the landforms were developed mostly during the late Quaternary (GRG and HG, 2002). Presence of broad and well-developed natural levee deposits associated with abandoned channels suggests that this region was fluvially very active in the recent past (GRG and HG, 2002). However, distributions of sand bodies in the study area are rather sparse, and are generally associated with seasonally active riverbeds and adjoining floodplains. Floodplains are distributed uniformly all over this study area (Fig. 2b). However, the floodplains of the western side are associated with some backswamps. These swampy lands are rejuvenated during the monsoon when the adjacent rivers are overflowed due to seasonal floods (GRG and HG, 2002). Moreover, numerous isolated smaller water bodies are located on the floodplains, mostly in the middle

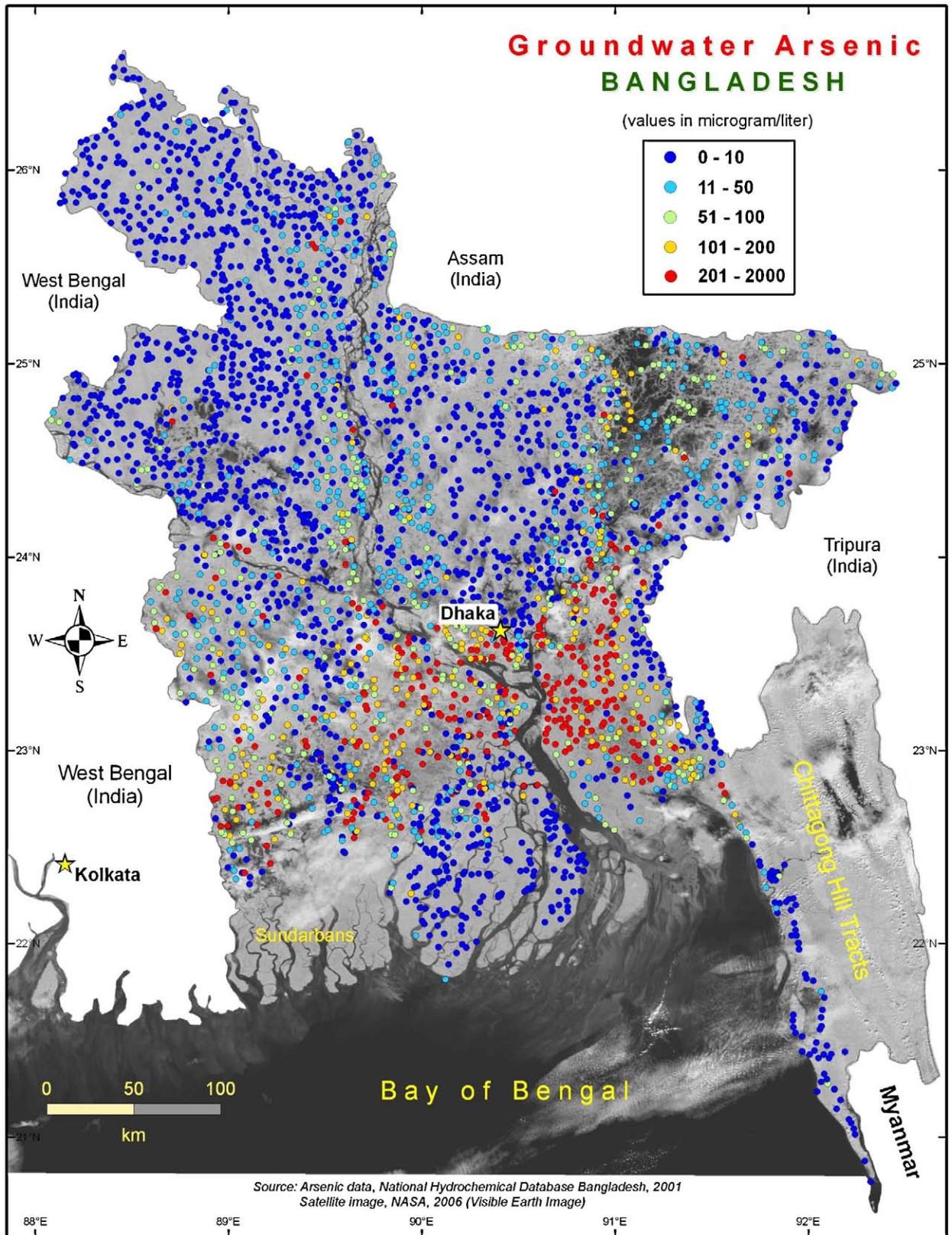


Fig. 1. Map of groundwater arsenic concentration in Bangladesh. Tubewell arsenic concentrations were collected from the National Hydrochemical Survey (NHS) of Bangladesh (BCS and DPHE, 2001). High arsenic (>50 $\mu\text{g/L}$) tubewells are mainly concentrated in the south-central parts of the country.

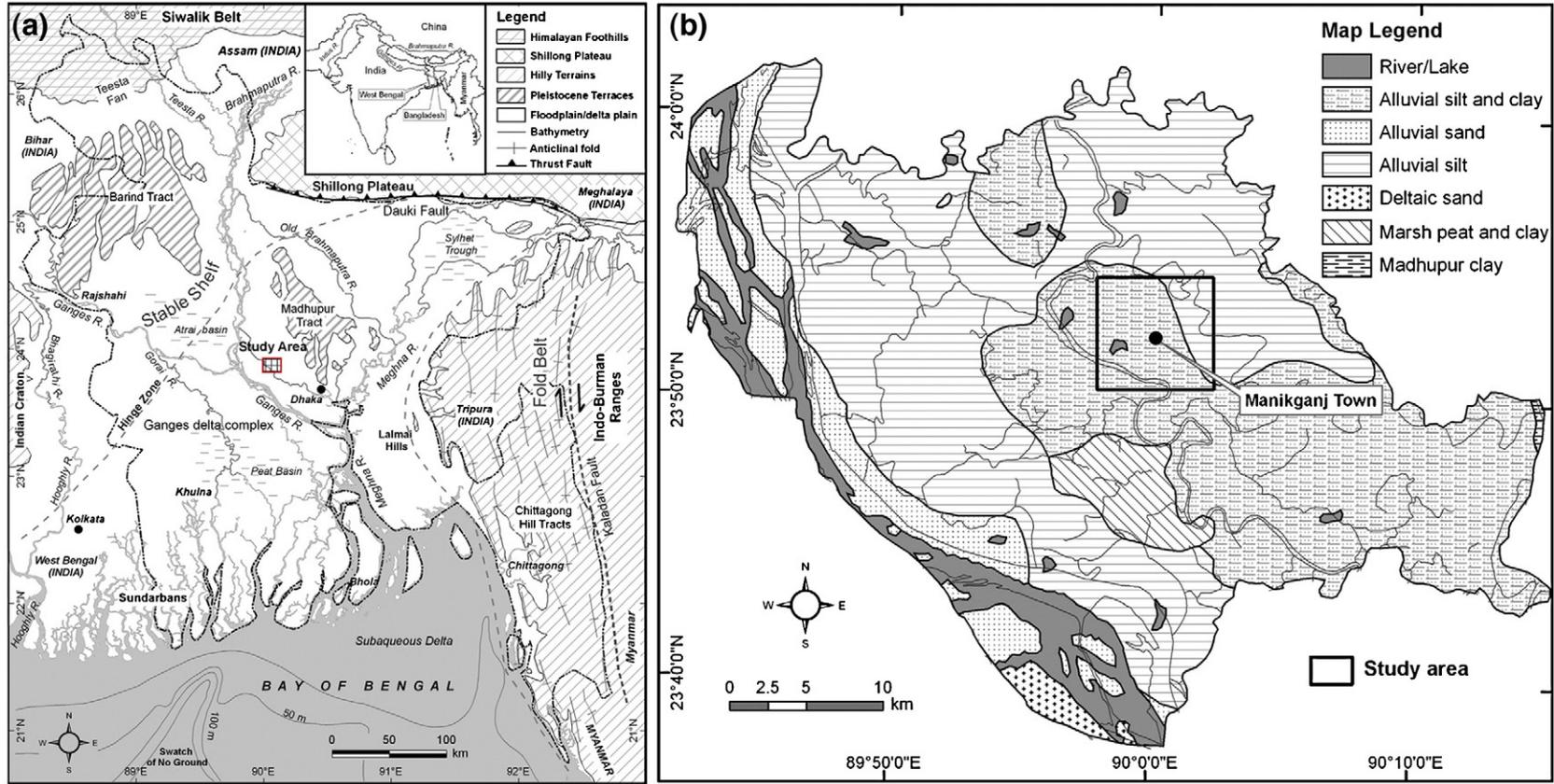


Fig. 2. (a) Simplified geological map of the Bengal Basin. Ganges, Brahmaputra and Meghna rivers form one of the largest deltaic systems, occupied by Bangladesh and West Bengal, India. Manikganj study area is shown as a rectangle, located to the southwest of Madhupur Tract; (b) Simplified surface geological map of the Manikganj district area. The study area is located within Manikganj town and adjoining areas, which are covered with alluvial silt and clay deposits.

portion of the study area, where several abandoned channels and natural levees form a complex landscape.

Several fining-upward sedimentary sequences formed the subsurface geology of the study area. Information from two drill holes reveals the stratigraphy and sedimentary history of

Manikganj and adjoining areas (Fig. 3). A description of sediment core samples, drill-site characterization and timing can be found in the following section on sampling and analytical procedures. Alluvial sediments ranging in size from clay to gravel formed the sedimentary sequences. Color of sediments varies from dark-

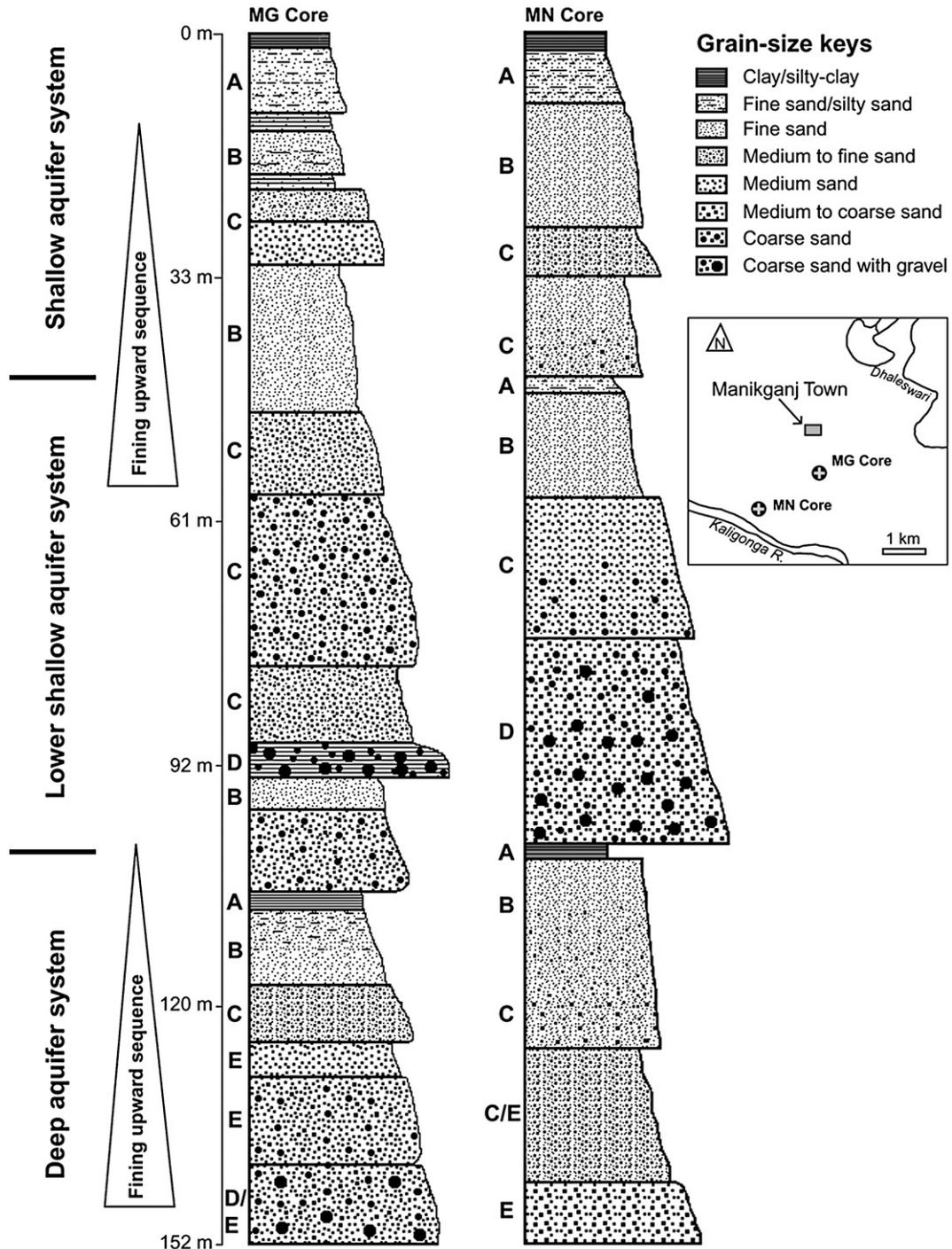


Fig. 3. Stratigraphy of the Quaternary sediments in the Manikganj area as revealed from core samples at two drilled core locations, MG and MN as shown on the index map on right-hand side. Two distinct fining-upward sequences are observed in both lithologies. Sedimentary facies: A – gray silt and silty clay, B – gray fine sand, C – gray to yellow medium to coarse sand, D – gravel-rich clean sand, E – orange-brown oxidized sand. Groundwater aquifer systems are developed within the fining-upward sedimentary sequences at Manikganj area.

gray (chemically reduced) to orange-brown (chemically oxidized) as noticed in many other fluvial-deltaic sedimentary sequences in Bangladesh (e.g., BGS and DPHE, 2001).

The uppermost fining-upward sequence is approximately 33 m thick in both drilling sites within the Manikganj town. The stratigraphy of this uppermost sequence at the “MG core” site is slightly different than the “MN core”, which is located in the natural levees of the Kaligonga river (Fig. 3). Presence of several clay and silty clay layers interbedded with sand in the MG core suggests a typical floodplain alluvial deposit formed by a meandering river. Some sand and silt layers contain organic matter (e.g., plant debris). Gray micaceous silt and clays within this sequence were probably deposited by meandering channels. Basal medium sand with occasional coarse sands with some plant debris and mica were deposited within the active meandering channels in the study area. Lithological characteristics and depositional patterns suggest a highstand sediment cycle, which is also seen in other locations of the country (BGS and DPHE, 2001).

The middle fining-upward sequence is very distinct in both well sites. The thickness of this sequence is approximately 55–60 m starting with coarse sand with gravels in MN core, but gravels in clay layer in MG core site (Fig. 3). The top of this fining-upward sequence is mostly fine sand. However, in MN site, there is a silty sand layer at 45 m deposited on top of fine sand suggesting a change in depositional energy for a short time. The bottom of this middle fining-upward sequence is characterized by gravel-rich detritus, although the gravel-rich layer is significantly thick (30–35 m) at the MN site, which is located closer to the Kaligonga river. The gravel-rich layer at the MG drill hole is thin (~6 m) and embedded within dark-gray clay. Gravels in both locations are mostly sub-rounded to rounded fragments of chert, quartzite and granite. This middle fining-upward sequence (45–90 m) can be interpreted as a lowstand to transgressive deposit recognized in other areas of the country. Based principally on lithological characteristics this middle stratigraphic unit can be correlated with the deposits beneath the Brahmaputra (Jamuna) floodplain described as the Dhamrai Formation (Table 1; Davies, 1989).

The bottom fining-upward sequence (100–152 m) is also distinct in both core sites in Manikganj area (Fig. 3). This sedimentary sequence is different from the top and middle units both in physical properties and composition. Color of sediments in this unit varies from yellowish-brown to orange-

brown as opposed to gray to dark-gray in the younger sediments. The bottom of this sequence is composed of coarse to medium sands. These sediments are lithologically similar to the Pliocene–Pleistocene Dupi Tila sand (Goodbred and Kuehl, 2000), which serves as the best regional aquifer in Bangladesh (BGS and DPHE, 2001).

Aquifers in the Manikganj study area are formed mostly by Quaternary alluvial sands (GRG and HG, 2002; Arafin, 2003). Three major aquifer systems are formed by three fining-upward sedimentary sequences in Manikganj (Turner, 2006; Shamsudduha, 2007) that are comparable with the three-aquifer classification scheme originally proposed by UNDP (1982). Aquifer systems are separated from each other by aquitard units, which are formed by clay and silty clay deposits of variable thickness. The uppermost aquifer system starts at approximately 10 m below the surface and extends vertically down to a depth of 33 m, which is composed mainly of very fine sand, mixed with silt (Fig. 3). The average thickness of this upper shallow aquifer system is approximately 15–20 m, consisting of several inter-connected aquifers of various sediment types. Most of the high arsenic-contaminated tubewells are screened in this aquifer system.

The lower shallow aquifer system consists of several potential groundwater aquifers ranging in total thickness of 55–60 m. Clast sizes in this aquifer system vary from fine sand to medium sand to even coarse sand with gravels toward the bottom of this middle stratigraphic unit (Fig. 3). Sediment colors change from gray to light yellowish-brown towards the bottom, indicating a change in chemical environment from reducing to slightly oxidizing toward the bottom.

The deep aquifer system in the Manikganj study area extends below ~100 m with a major change in sediment composition and color. Most aquifers in this system are composed of fine to medium grained yellowish-brown to bright orange-brown sands, indicating highly oxidized chemical conditions in these aquifers. This system is similar as the “deeper aquifer” proposed by BGS and DPHE (2001) and is believed to yield excellent quality groundwater in most parts of Bangladesh.

3. Sampling and analytical procedures

3.1. Groundwater sampling

The present study has compiled geochemical results from 85 groundwater samples. Fifty-one groundwater samples (MK-

Table 1

Simplified stratigraphic succession of the upper Cenozoic deposits in Manikganj study area (modified after Davies, 1989)

Age	Stratigraphic unit	Subdivision in Manikganj area	Lithology and facies description	Thickness (meter)
Recent Holocene	Alluvium Dhamrai Formation	Upper fining-upward sequence	Fine sand, silt, silty clay and clay with micas and plat fragments Light-gray to dark-gray, fine to medium sand with silt and occasional clay. Some sand layers are rich in mica and dark-colored heavy minerals. Highstand fluvial deposits	5–10 20–30
		Middle fining-upward sequence	Fine to medium sand, with coarse sand and gravel toward the bottom of the sequence. Color of the sediments varies from gray to light yellowish-gray. Less organic matter in sediments. Lowstand System Tract to Transgressive Tract alluvial deposits	65–75
Pliocene– Pleistocene	Dupi Tila Formation	Lower fining-upward sequence	Yellowish-brown to orange-brown colored fine to medium sand with occasional gravel. Sediments are highly oxidized and iron-coatings are common on sand grains. Pre-Lowstand fluvial deposits	>50

Table 2
Major groundwater chemistry of surveyed tubewells in the Manikganj study area

Sample no.	Latitude (degree)	Longitude (degree)	Well depth (m)	Ca	Na	Mg	K	NO ₃	Cl	PO ₄	SO ₄	HCO ₃	Ionic balance (%)
MK-01	23.865	90.001	178.3	68.2	58.8	25.9	4.50	0.01	25.4	0.01	3.42	412.4	0.66
MK-02	23.865	90.001	45.0	88.1	16.2	32.5	3.81	0.01	7.1	0.01	6.35	439.2	0.34
MK-03	23.848	90.003	79.5	71.3	27.2	25.8	4.25	1.06	11.4	0.01	2.40	370.9	0.53
MK-04	23.851	90.004	–	64.7	22.7	24.0	4.19	4.43	10.9	0.01	1.86	334.3	0.47
MK-05	23.852	90.004	105.9	61.4	22.7	25.2	4.54	6.02	10.4	0.01	0.01	341.6	0.35
MK-06	23.853	90.004	100.0	65.1	19.9	21.6	3.87	5.26	10.6	0.01	0.01	336.7	0.17
MK-07	23.869	90.002	137.2	62.4	45.1	24.7	3.25	0.01	15.4	0.01	0.01	383.1	0.47
MK-08	23.873	90.009	228.6	54.4	35.0	20.8	3.08	0.01	15.5	0.01	1.89	297.7	0.67
MK-09	23.846	90.017	73.2	62.2	18.6	23.4	3.86	1.25	4.5	0.01	4.26	327.0	0.36
MK-10	23.848	90.015	36.6	71.7	13.2	32.2	2.76	0.01	17.9	0.01	26.22	339.2	0.27
MK-11	23.847	90.016	18.3	102.4	26.4	28.5	2.43	1.83	35.9	0.01	2.47	417.2	0.77
MK-12	23.850	90.015	73.2	67.6	15.3	28.5	3.27	0.01	14.2	0.01	0.01	339.2	0.50
MK-13	23.846	90.022	36.6	82.1	17.6	30.2	7.23	4.45	7.3	0.01	8.52	388.0	0.79
MK-14	23.845	90.023	27.4	75.2	18.1	34.5	13.97	0.01	10.5	0.01	40.00	363.5	0.65
MK-15	23.846	90.023	73.2	61.3	13.7	21.0	4.21	4.05	4.3	0.01	3.35	307.4	0.26
MK-16	23.866	89.998	22.9	78.8	46.3	26.4	39.97	28.88	58.5	5.32	49.92	312.2	1.34
MK-17	23.867	89.998	15.2	71.6	14.8	20.9	8.67	0.01	23.3	6.74	12.62	263.5	0.92
MK-18	23.872	89.996	73.2	67.6	25.2	24.4	3.36	0.01	15.0	0.01	0.01	317.2	0.94
MK-19	23.873	89.996	36.6	85.5	16.7	25.9	3.72	0.01	19.3	0.01	0.01	351.4	0.92
MK-20	23.873	89.996	18.3	68.6	12.8	21.8	3.09	0.01	12.2	0.01	1.84	314.8	0.32
MK-21	23.853	90.030	73.2	58.8	14.3	24.7	3.65	0.01	21.3	0.01	43.00	322.1	-1.09
MK-22	23.852	90.026	22.9	58.0	8.2	21.6	2.98	0.01	4.0	0.01	0.01	295.2	0.15
MK-23	23.852	90.027	36.6	68.4	10.8	21.2	3.62	0.01	4.0	0.01	0.01	317.2	0.41
MK-24	23.856	89.983	73.2	56.9	11.7	19.9	4.27	0.01	4.9	0.01	0.01	270.9	0.51
MK-25	23.854	89.982	36.6	71.4	10.7	21.3	3.03	0.01	5.5	0.01	0.01	348.9	-0.02
MK-26	23.858	90.032	22.9	116.1	12.5	30.3	3.23	0.01	61.5	0.01	17.65	370.9	0.73
MK-27	23.857	90.033	36.6	57.6	12.8	25.7	4.23	0.01	5.3	0.01	1.96	329.4	0.07
MK-28	23.861	90.033	73.2	71.4	16.4	25.0	3.64	0.01	6.2	0.01	0.01	368.4	0.21
MK-29	23.858	90.034	15.2	71.9	11.2	20.0	3.67	0.69	11.7	0.01	0.01	290.4	0.73
MK-30	23.863	90.015	21.4	95.1	13.1	19.8	3.75	5.56	34.0	0.01	41.70	273.3	0.73
MK-31	23.864	90.015	18.3	51.0	12.8	19.6	12.21	1.04	4.0	3.26	14.32	244.0	0.62
MK-32	23.860	90.017	37.2	55.7	7.5	23.5	2.41	0.01	4.2	0.01	22.34	263.5	0.20
MK-33	23.867	90.025	73.2	56.3	11.9	22.6	28.63	1.14	7.4	0.01	0.01	305.0	0.70
MK-34	23.867	90.025	22.9	91.1	21.6	43.7	4.38	4.61	71.0	0.01	73.42	341.6	0.06
MK-35	23.869	90.025	45.7	58.1	13.8	23.9	18.36	0.01	6.9	0.01	12.41	307.4	0.45
MK-36	23.857	89.996	73.2	70.4	11.7	22.6	6.08	0.01	20.7	0.01	12.33	290.4	0.43
MK-37	23.858	89.996	36.6	73.6	10.9	27.8	10.20	0.01	7.2	0.01	13.91	348.9	0.48
MK-38	23.855	90.000	21.3	135.2	18.2	34.5	0.67	0.01	21.5	0.01	41.68	483.1	1.00
MK-39	23.863	89.998	45.7	93.5	10.5	40.1	5.62	0.01	36.3	0.01	18.87	397.7	0.63
MK-40	23.872	90.017	73.2	62.0	12.7	18.6	3.73	0.01	5.0	0.01	1.81	297.7	0.88
MK-41	23.872	90.017	18.3	72.9	11.5	29.0	3.91	0.01	2.6	0.01	19.12	322.1	0.72
MK-42	23.869	90.019	37.2	47.8	19.0	28.1	20.83	0.01	6.2	0.01	1.87	312.3	0.88
MK-43	23.844	90.027	73.2	86.2	22.0	43.9	4.26	0.01	34.2	0.01	47.17	375.8	-1.76
MK-44	23.844	90.027	45.7	51.5	18.9	28.6	21.40	0.01	52.2	0.01	75.73	305.0	3.22
MK-45	23.844	90.022	18.3	125.4	22.0	38.5	1.81	0.01	6.9	0.01	4.20	422.1	5.00
MK-46	23.841	89.987	202.7	143.0	18.9	33.8	0.82	0.01	1.4	0.01	1.85	336.7	1.02
MK-47	23.872	89.974	192.0	41.7	36.1	20.1	3.66	0.01	4.1	0.01	2.90	256.2	1.01
MK-48	23.877	89.986	185.9	58.7	31.4	21.6	3.32	3.34	5.4	0.01	0.01	305.0	1.32
MK-49	23.879	89.996	185.9	64.3	36.2	28.8	3.01	0.01	9.8	0.01	1.76	341.6	0.94
MK-50	23.878	90.021	195.1	54.3	56.1	21.0	3.09	0.01	13.3	0.01	2.25	341.6	0.90
MK-51	23.861	90.001	137.2	39.3	98.8	16.0	3.32	0.01	32.6	0.01	1.73	353.8	0.66
MK-52	23.880	89.992	22.9	49.2	15.2	20.4	3.69	0.02	5.5	0.05	0.06	268.4	0.33
MK-60	23.880	89.992	25.0	63.4	13.3	27.7	2.96	0.02	5.7	0.04	0.06	302.6	0.98
MK-61	23.881	89.992	15.0	108.0	12.5	30.7	1.70	0.02	22.6	0.04	1.86	396.5	1.33
MK-62	23.857	89.975	39.6	83.5	19.2	29.4	7.98	0.02	4.8	0.04	4.19	388.0	1.04
MK-63	23.857	89.975	22.9	111.0	18.2	40.5	5.83	0.02	12.0	0.04	9.27	481.9	1.38
MK-64	23.857	89.976	24.4	84.0	14.0	24.5	2.99	0.02	9.4	0.04	0.06	366.0	0.63
MK-65	23.881	89.992	21.3	82.6	19.2	30.9	4.10	0.02	33.7	0.04	0.06	357.5	0.79
MK-66	23.881	89.992	18.3	133.0	27.6	45.5	2.48	0.19	42.9	0.04	18.30	463.6	2.45
MK-67	23.881	89.993	9.2	118.0	16.4	29.0	6.92	0.06	16.9	0.04	9.90	409.9	1.76
MK-68	23.874	89.993	29.0	120.0	31.6	35.9	5.39	0.02	58.8	0.04	26.20	429.4	1.21
MK-69	23.873	89.993	73.2	78.2	36.7	27.1	4.14	0.02	11.5	0.02	0.06	394.1	1.05
MK-70	23.879	89.993	15.9	87.9	14.9	27.6	2.78	0.01	18.4	0.02	0.03	396.5	0.36
MK-71	23.883	89.992	15.2	73.5	11.2	21.7	3.36	0.01	10.7	0.02	0.03	342.8	0.10
MK-72	23.886	89.989	29.6	54.5	15.4	34.0	28.00	0.01	16.8	0.02	26.40	351.4	-0.60
MK-73	23.886	89.989	48.8	50.0	18.4	22.6	8.10	0.02	14.5	0.02	1.10	291.6	0.15
MK-74	23.860	89.997	16.8	107.0	15.1	27.2	0.67	0.02	22.2	0.03	2.30	444.1	0.30
MK-75	23.856	89.984	15.9	96.1	15.5	46.1	38.00	0.02	39.4	0.03	12.20	553.9	-1.18

Table 2 (continued)

Sample no.	Latitude (degree)	Longitude (degree)	Well depth (m)	Ca	Na	Mg	K	NO ₃	Cl	PO ₄	SO ₄	HCO ₃	Ionic balance (%)
MK-76	23.886	89.981	15.9	105.0	19.9	21.7	4.38	0.01	27.2	0.02	17.00	407.5	0.20
MK-77	23.884	89.976	27.4	82.6	16.1	22.3	4.00	0.02	10.0	0.02	0.03	381.9	0.22
MK-78	23.902	89.988	61.0	126.0	17.9	29.1	1.87	0.02	9.0	0.03	25.20	500.2	0.53
MK-79	23.902	89.988	15.9	63.2	17.8	19.7	4.50	0.01	4.4	0.02	0.03	336.7	0.02
MK-80	23.888	89.990	7.6	118.0	24.4	26.8	4.10	0.02	30.8	0.03	8.35	485.6	0.26
MK-81	23.880	89.992	12.2	85.2	24.7	31.9	3.55	0.02	17.1	0.03	0.91	438.0	0.36
MK-82	23.888	89.989	15.9	74.1	20.1	40.0	7.22	0.02	30.0	0.03	8.15	420.9	0.13
MK-83	23.894	89.995	19.8	67.6	12.2	31.3	4.03	0.01	12.9	0.02	22.80	341.6	0.14
MK-84	23.890	89.995	15.2	41.1	16.1	13.3	2.72	0.01	5.9	0.02	10.90	266.0	-0.84
MK-85	23.888	89.994	22.9	98.3	14.2	21.4	4.00	0.01	4.9	0.02	11.70	417.2	0.17
MK-86	23.882	89.994	21.3	71.4	17.4	25.5	3.03	0.01	15.3	0.02	19.10	335.5	0.17
MK-87	23.876	89.996	35.1	69.8	20.3	25.9	2.76	0.01	9.0	0.02	5.80	363.6	0.24
MK-88	23.874	89.994	13.7	76.2	25.0	22.4	5.83	0.01	12.3	0.02	0.03	378.2	0.34
MK-89	23.873	89.979	16.8	60.8	17.5	21.8	10.70	0.12	12.5	0.23	16.20	307.4	0.13
MK-90	23.873	89.970	24.4	32.1	7.8	19.7	2.38	0.02	5.1	0.01	6.65	181.8	0.36
MKIW-01	23.880	89.992	24.4	52.9	13.9	21.8	2.51	0.01	5.4	0.01	0.03	295.2	0.11
MKIW-02	23.874	89.993	36.6	74.6	28.3	21.6	15.80	-	-	-	-	-	-

Concentrations of all the major elements are expressed in mg/L.

01–MK-51) were collected in 2001 (McCarthy, 2001; Arafin, 2003). Additional 11 water samples (MK-52, and MK-60–MK-69) were collected by in 2005 (Turner, 2006). Remaining 23 water samples (MK-70–MK-90, and MKIW-01–MKIW-02) were collected between December 2005 and January 2006 (Shamsudduha, 2007). At each sampling site, the geographic location was recorded by a hand-held Global Positioning System (GPS), and information on well depth was acquired through personal communication with tubewell owners. At each site, tubewell was purged for approximately 5–10 min (depending on depths to well screen) to expel any standing water into the well pipes and water was pumped into a plastic beaker in which the field parameters (temperature, oxidation–reduction potential (ORP), pH, H₂S, dissolved oxygen (DO) and specific conductance) were measured using hand-held portable instruments. Raw water samples were collected into two distilled water washed 60 mL plastic bottles, which included: (i) one filtered (Nalgene 0.45 µm filter) sample for anion analysis; (ii) one filtered (Nalgene 0.45 µm filter) and acidified sample with concentrated nitric acid (HNO₃) for analysis of cations and trace elements, including As. Anion concentrations for 34 water samples (MK-52, MK-60–MK-69, MK-70–MK-90, and MKIW-01–MKIW-02) were measured using ion chromatography (IC), but for 51 water samples (MK-01–MK-51), the Genesys-5 spectrophotometer and titration method were used. Cation concentrations were measured using inductively coupled plasma-mass spectrometry (ICP-MS) and inductively coupled plasma-optical emission spectrometer (ICP-OES) methods for above-mentioned 34 water samples, but for samples MK-01 to MK-51, the flame photometer and titration methods were used. In all cases, the trace elements, including As, Fe, and Mn in groundwater samples, were analyzed at the Activation Laboratories in Ontario, Canada, using ICP-MS and ICP-OES methods.

3.2. Drilling and sediment sampling

Sediment core samples from two boreholes in the study area (shown in Fig. 3) were collected by a split-spoon sampler made from a 0.6-m (2 ft) section of PVC pipe (ID 5 cm) with a rotary drill rig operated by the Bangladesh Water Development Board

(BWDB). Two drill-cores were sampled in two different time periods. Most sediment core samples used in this study were collected from a recently drilled site in Manikganj between 25th December 2005 and 8th January 2006. This newly collected drill-core sampling location is marked as MN core site (Fig. 3). The length of the drilled core was 152 m, and a total of 100 core samples were collected with a continuous core recovery from the top 30 m, and discontinuously (sampling intervals 3 m from 30 m to 90 m, and 6 m from 100 m to 152 m) collected to the bottom of the borehole. The core samples were collected in plastic PVC tubes with a maximum length of 60 cm, but the average length was about 30 cm. The ends of tubes containing sediment were wax-sealed on site immediately after recovery from the drill hole and completion of brief notes on sediment color and texture. Another set of sediment core samples (47 samples) that were collected in January 2001 from another site in Manikganj Town (MG core site as shown in Fig. 3) by GRG and HG (2002) has been included in this study. The MG core samples were collected following the same procedure as described for the MN core samples. Wax-sealed core samples (a total of 47 samples) in heavy-duty PVC tubes were preserved in a chemical laboratory at the Geology Department of Dhaka University at ambient temperature and shipped to the Himalayan Research Laboratory (HRL) at Auburn University for further processing and analysis.

3.3. Sediment geochemistry analysis

A total of 32 samples were selected from both the MN and MG sediment cores based on the information on groundwater arsenic concentrations at different depths in the study area. Frequency of sediment sampling for geochemical analysis was primarily based on the occurrences of higher arsenic concentration in groundwaters. Mostly finer sediments were targeted for whole rock geochemical analysis since studies shown that arsenic concentration is high in fine-grained sediments (BCS and DPHE, 2001; Horneman et al., 2004). Samples were dried in an oven at about 50 °C for approximately 24 h. Approximately 20 g of dried sediment for each sample was crushed with a mortar and pestle. Powdered sediment samples were sent to the ACME Laboratories Ltd. in Vancouver, Canada for analysis. In

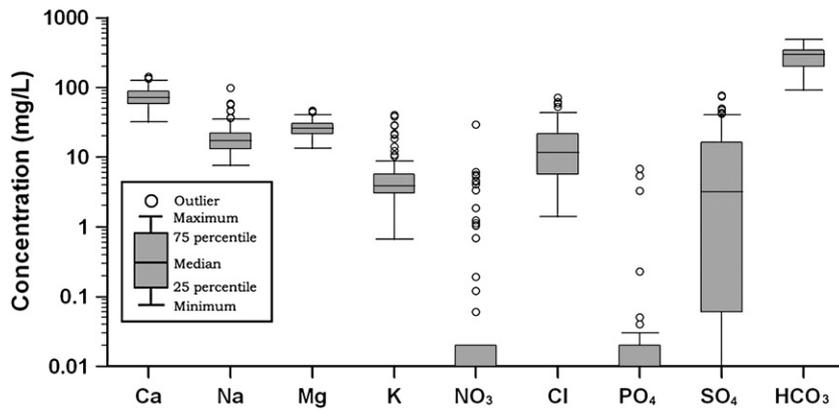


Fig. 4. Box plot showing variations of the major groundwater ion concentrations in the Manikganj study area. The inset diagram is shown as the key to the box plot.

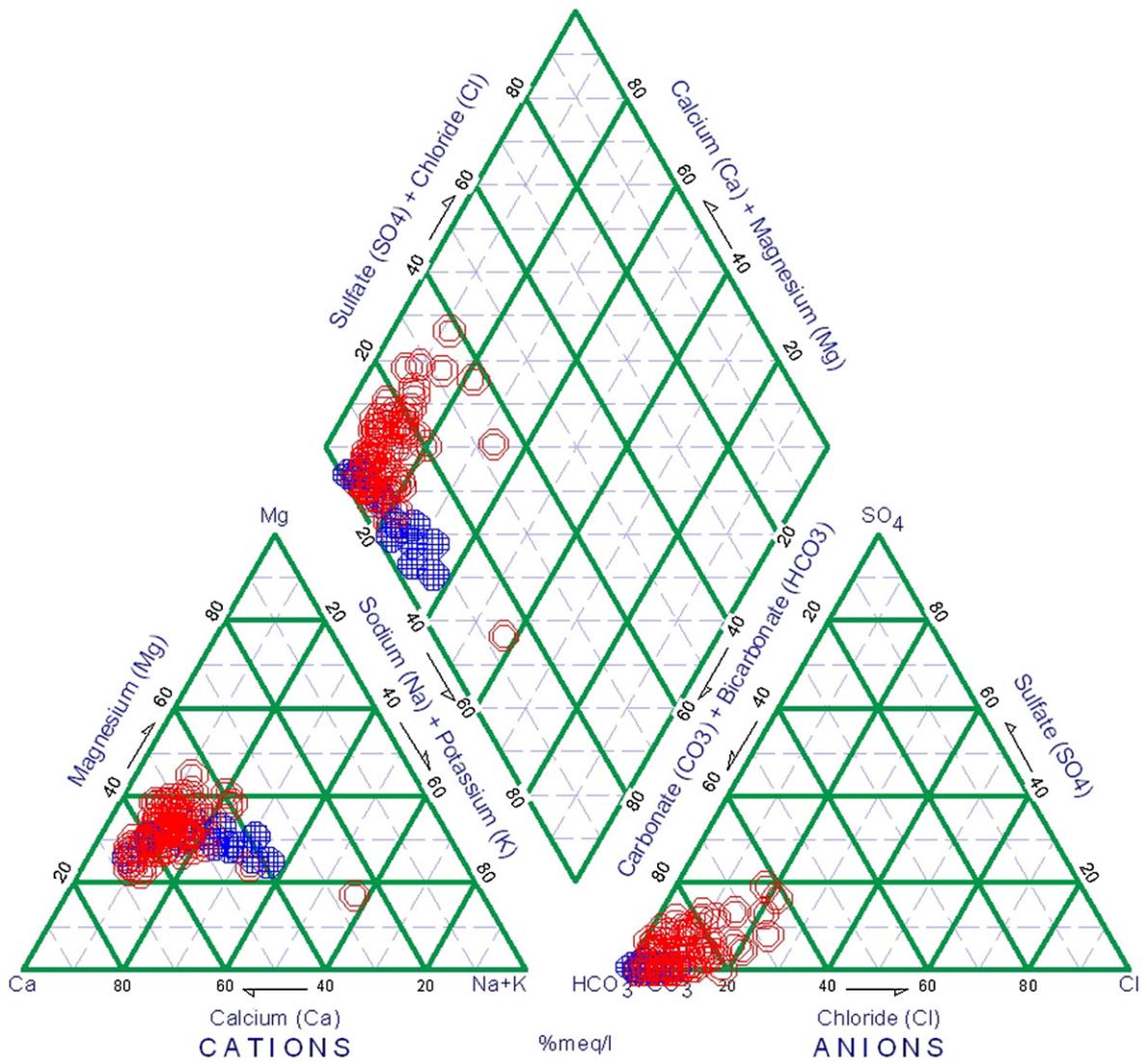


Fig. 5. Piper diagram illustrating the main hydrochemical features of Manikganj groundwater. Shallow (<100 m; open circle) and deep (>100 m; gridded circle) major groundwater compositions are slightly different in the study area. The overall groundwater type is Ca-HCO₃, which is consistent with groundwater compositions in most alluvial aquifers in Bangladesh.

Table 3

Trace elements concentrations in groundwater of the surveyed tubewells in the Manikganj study area

Sample no.	Depth (m)	Fe (mg/L)	As ($\mu\text{g/L}$)	Mn (mg/L)	Sr (mg/L)	Zn ($\mu\text{g/L}$)	Al (mg/L)	Ba (mg/L)	Si (mg/L)
MK-01	178.3	4.59	8.4	0.87	0.68	0.22	0.01	0.13	n.m.
MK-02	45.0	17.74	31.2	0.26	0.39	0.14	0.03	0.16	n.m.
MK-03	79.5	9.11	43.9	0.29	0.33	0.02	0.01	0.13	n.m.
MK-04	–	0.07	17.6	0.12	0.30	0.18	0.01	0.08	n.m.
MK-05	105.9	7.32	46.9	0.22	0.30	0.02	0.02	0.11	n.m.
MK-06	100.0	7.72	64.8	0.28	0.29	0.01	0.02	0.16	n.m.
MK-07	137.2	0.77	33.1	0.75	0.40	0.02	0.02	0.16	n.m.
MK-08	228.6	0.90	4.1	0.69	0.38	0.03	0.01	0.10	n.m.
MK-09	73.2	11.60	18.1	0.61	0.24	0.03	0.02	0.17	n.m.
MK-10	36.6	14.82	3.6	0.20	0.24	0.02	0.01	0.19	n.m.
MK-11	18.3	14.35	28.4	1.60	0.32	0.02	0.03	0.09	n.m.
MK-12	73.2	15.95	22.3	0.59	0.28	0.11	0.01	0.16	n.m.
MK-13	36.6	7.54	17.1	0.53	0.35	0.02	0.02	0.24	n.m.
MK-14	27.4	13.15	14.1	1.23	0.20	0.03	0.01	0.26	n.m.
MK-15	73.2	10.37	59.5	0.33	0.27	0.05	0.00	0.16	n.m.
MK-16	22.9	0.16	4.0	0.04	0.12	0.02	0.01	0.06	n.m.
MK-17	15.2	0.25	4.8	0.54	0.13	0.01	0.02	0.04	n.m.
MK-18	73.2	6.55	43.5	0.21	0.34	0.01	0.02	0.17	n.m.
MK-19	36.6	10.16	105.9	0.36	0.42	0.03	0.01	0.19	n.m.
MK-20	18.3	8.67	102.0	0.35	0.33	0.01	0.02	0.14	n.m.
MK-21	73.2	9.27	55.3	0.51	0.26	0.02	0.01	0.15	n.m.
MK-22	22.9	16.54	35.3	1.43	0.24	0.02	0.02	0.11	n.m.
MK-23	36.6	13.25	69.4	0.30	0.29	0.01	0.01	0.15	n.m.
MK-24	73.2	7.95	78.6	0.41	0.24	0.02	0.01	0.13	n.m.
MK-25	36.6	26.98	60.2	0.52	0.21	0.04	0.02	0.20	n.m.
MK-26	22.9	2.23	12.2	4.16	0.37	0.02	0.03	0.11	n.m.
MK-27	36.6	18.92	51.2	0.66	0.26	0.01	0.03	0.15	n.m.
MK-28	73.2	15.13	27.6	0.71	0.29	0.01	0.02	0.20	n.m.
MK-29	15.2	0.71	5.8	1.12	0.23	0.01	0.13	0.09	n.m.
MK-30	21.4	0.21	1.2	0.60	0.22	0.02	0.02	0.09	n.m.
MK-31	18.3	0.19	3.9	0.44	0.06	0.01	0.02	0.06	n.m.
MK-32	37.2	10.55	9.8	0.87	0.20	0.02	0.01	0.09	n.m.
MK-33	73.2	2.46	0.8	0.66	0.23	0.01	0.02	0.25	n.m.
MK-34	22.9	8.06	16.4	0.94	0.29	0.02	0.02	0.20	n.m.
MK-35	45.7	0.00	9.4	0.88	0.22	0.01	0.01	0.18	n.m.
MK-36	73.2	6.67	63.7	0.28	0.27	0.01	0.03	0.15	n.m.
MK-37	36.6	5.69	38.9	1.04	0.25	0.02	0.01	0.20	n.m.
MK-38	21.3	0.00	2.3	0.94	0.31	0.02	0.02	0.09	n.m.
MK-39	45.7	0.60	58.8	0.76	0.42	0.01	0.00	0.14	n.m.
MK-40	73.2	0.01	n.m.	0.88	0.25	0.31	0.00	0.16	n.m.
MK-41	18.3	0.16	24.0	2.05	0.28	0.01	0.02	0.09	n.m.
MK-42	37.2	0.00	11.1	0.75	0.14	0.02	0.02	0.17	n.m.
MK-43	73.2	0.03	4.1	0.90	0.29	0.01	0.03	0.14	n.m.
MK-44	45.7	7.79	21.2	0.79	0.15	0.02	0.01	0.20	n.m.
MK-45	18.3	0.19	14.9	3.36	0.38	0.03	0.03	0.20	n.m.
MK-46	202.7	3.51	0.3	1.23	0.31	0.03	0.00	0.12	n.m.
MK-47	192.0	0.40	9.2	0.38	0.28	0.03	0.01	0.12	n.m.
MK-48	185.9	0.32	0.4	0.73	0.37	0.03	0.04	0.14	n.m.
MK-49	185.9	0.00	0.3	0.89	0.44	0.02	0.03	0.16	n.m.
MK-50	195.1	0.00	0.5	0.63	0.37	0.07	0.02	0.16	n.m.
MK-51	137.2	0.00	8.9	0.31	0.26	0.42	0.01	0.12	n.m.
MK-60	25.0	0.13	12.3	0.37	0.32	0.00	0.00	0.06	9.4
MK-61	15.0	0.13	8.8	0.00	0.34	0.01	0.00	0.07	7.7
MK-62	39.6	0.32	5.2	0.95	0.33	0.02	0.00	0.13	9.9
MK-63	22.9	0.15	6.5	0.00	0.29	0.00	0.00	0.10	11.7
MK-64	24.4	1.15	6.1	0.44	0.20	0.06	0.00	0.11	13.1
MK-65	21.3	0.15	11.1	0.00	0.39	0.00	0.00	0.07	15.9
MK-66	18.3	0.19	4.4	0.00	0.51	0.00	0.00	0.11	16.6
MK-67	9.2	0.15	4.8	0.00	0.36	0.02	0.00	0.10	13.8
MK-68	29.0	0.49	58.1	0.36	0.50	0.00	0.00	0.18	22.9
MK-69	73.2	0.26	20.8	0.23	0.32	0.02	0.00	0.14	20.8
MK-52	22.9	0.25	12.5	0.49	0.26	0.00	0.00	0.06	17.1
MK-70	15.9	8.86	118.0	0.55	0.44	0.04	0.00	0.14	22.5
MK-71	15.2	6.65	163.0	1.73	0.41	0.00	0.00	0.14	19.6
MK-72	29.6	5.74	15.9	0.68	0.20	0.02	0.00	0.31	22.3
MK-73	48.8	7.06	47.7	0.75	0.23	0.10	0.00	0.22	22.8
MK-74	16.8	2.27	2.9	0.92	0.29	0.00	0.00	0.05	13.7

(continued on next page)

Table 3 (continued)

Sample no.	Depth (m)	Fe (mg/L)	As ($\mu\text{g/L}$)	Mn (mg/L)	Sr (mg/L)	Zn ($\mu\text{g/L}$)	Al (mg/L)	Ba (mg/L)	Si (mg/L)
MK-75	15.9	9.71	14.4	0.30	0.34	0.00	0.00	0.04	23.5
MK-76	15.9	4.85	20.2	1.30	0.23	0.03	0.00	0.05	19.9
MK-77	27.4	0.13	16.2	0.61	0.23	0.00	0.00	0.12	21.5
MK-78	61.0	12.40	20.0	0.49	0.32	0.02	0.00	0.16	19.8
MK-79	15.9	9.47	54.8	0.85	0.30	0.00	0.00	0.19	23.7
MK-80	7.6	0.20	1.7	2.24	0.32	0.34	0.01	0.08	23.8
MK-81	12.2	13.40	78.4	0.70	0.45	0.01	0.00	0.18	24.8
MK-82	15.9	3.51	8.6	0.28	0.22	0.00	0.00	0.02	22.7
MK-83	19.8	0.59	41.5	0.85	0.17	0.00	0.00	0.08	21.7
MK-84	15.2	0.10	9.6	0.58	0.15	0.00	0.00	0.69	18.5
MK-85	22.9	1.83	5.6	0.28	0.18	0.01	0.00	0.15	19.9
MK-86	21.3	12.70	63.9	1.18	0.27	0.01	0.00	0.16	23.3
MK-87	35.1	4.32	11.9	0.56	0.21	0.00	0.00	0.13	21.5
MK-88	13.7	6.92	184.0	0.36	0.33	0.00	0.00	0.16	25.8
MK-89	16.8	0.47	39.0	1.05	0.32	0.05	0.00	0.80	20.6
MK-90	24.4	1.61	40.3	0.65	0.08	0.01	0.00	0.05	24.5
MKIW-01	24.4	5.62	91.4	0.38	0.27	0.00	0.00	0.01	21.1
MKIW-02	36.6	7.88	191.0	0.27	0.24	12.60	0.01	0.04	22.8

n.m. – not measured.

the lab, 0.50 g sample was leached with 3 mL 2–2–2 HCl–HNO₃–H₂O at 95 °C for 1 h and diluted to 10 mL, and analyzed with the inductively coupled plasma-mass spectrometry (ICP-MS) method. A total of 36 major and trace elements were analyzed from the selected 32 sediment samples.

3.4. Petrographic analysis

Petrographic analysis was performed with a petrographic microscope (Nikon model no. E600 POL) attached with an automatic point-counter and a photomicrographic setup. Sixteen thin-sections were prepared from core sands at different depth intervals. Thin-sections were stained for both potassium and plagioclase feldspars.

Modal analyses were conducted following the Gazzi–Dickinson method, whereby sand-sized minerals within lithic fragments are counted as individual minerals in order to normalize for grain-size variation (Dickinson, 1970; Ingersoll et al., 1984). A minimum of 250 points were counted per sample. However, up to 300 framework points were counted for the samples that had greater compositional diversity. Modal sandstone compositions have been plotted on standard ternary

diagrams (QtFL – total quartz, total feldspar, and lithic fragments; QmFLt – monocrySTALLINE quartz, total feldspar, and total lithic fragments, QmPK – monocrySTALLINE quartz, plagioclase feldspar, and potassium feldspar, and LsLvLm – sedimentary lithic, volcanic lithic, and metamorphic lithic fragments) to assess temporal changes in provenance (Dickinson, 1970).

Heavy mineral assemblages from 16 sediment samples were separated by gravity settling method with tetrabromoethane (Br₂CHCHBr₂, density 2.89–2.96 g/cm³) (Uddin and Lundberg, 1998b). Dry and weighed samples were added to the heavy liquid in a separating funnel for 24 h. The mixture was stirred periodically to ensure that the grains were thoroughly wetted. Heavy minerals then gradually accumulated at the bottom of the funnel above the pinch clip. When sinking of heavy minerals stopped, the stopcock was opened slowly, and heavy fractions were allowed to pour into filter paper at the lower funnel. The stopcock was then closed immediately to leave a layer of clear liquid below the lighter fraction. The light fraction was then drained into a new funnel. Both fractions were washed thoroughly with acetone and put into an oven for drying. Heavy fractions were then

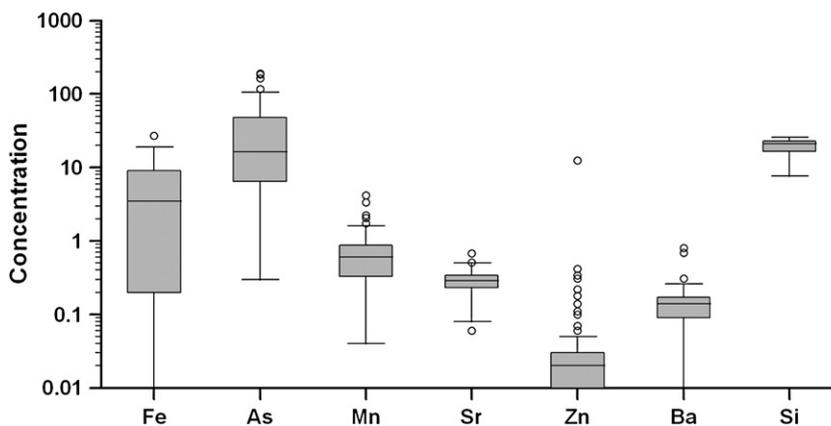


Fig. 6. Box plot showing variations of the minor groundwater ion concentrations in the Manikganj study area. The inset diagram in Fig. 4 is referred as the key to this plot. Concentrations of As and Zn are in $\mu\text{g/L}$, but for the other constituents, the concentrations are given in mg/L.

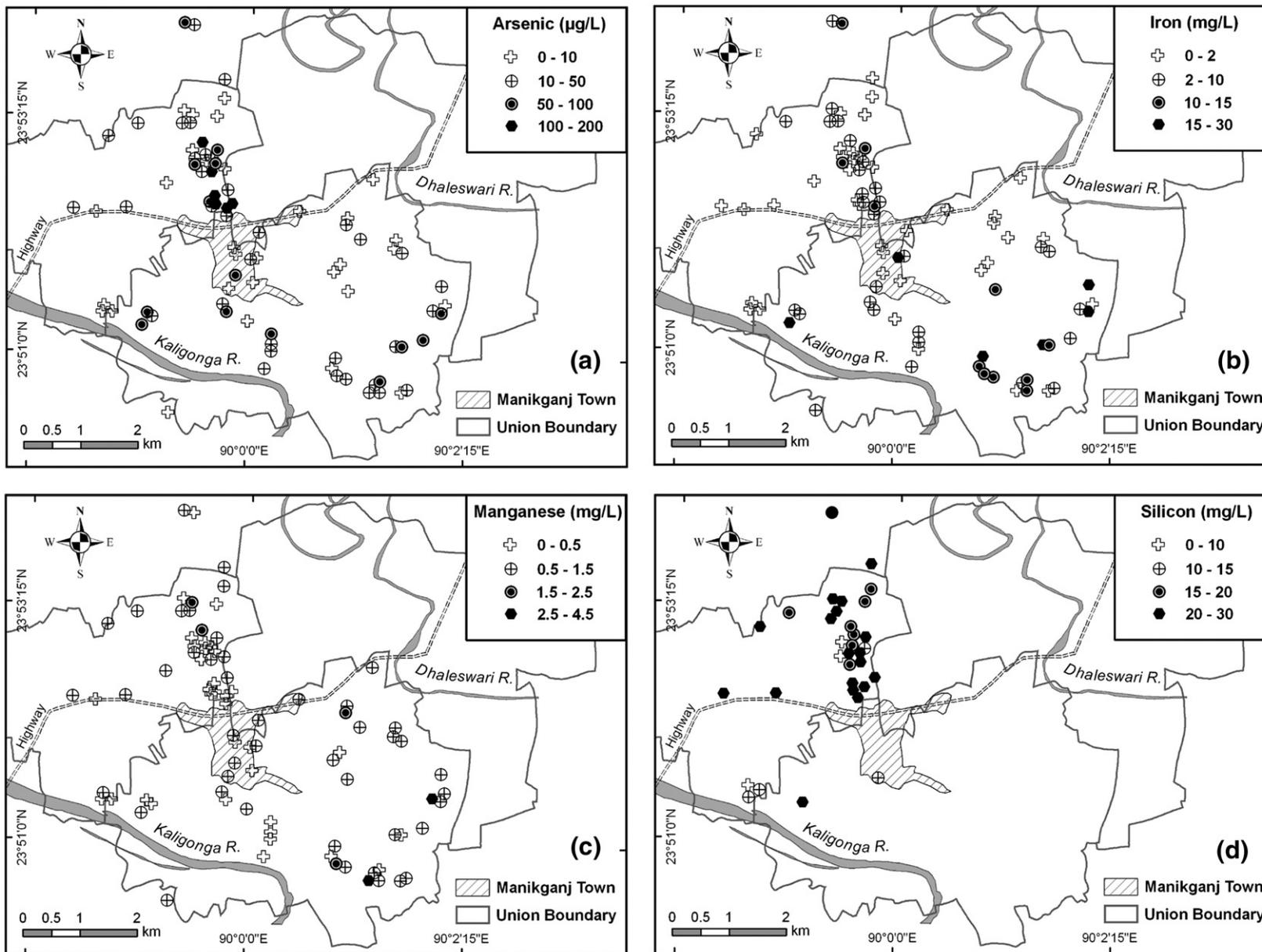


Fig. 7. Spatial distributions of dissolved arsenic (a) As, (b) Fe, (c) Mn, and (d) Si in the groundwater in Manikganj area. High As values are corresponding spatially with high concentrations of Fe, Mn and Si.

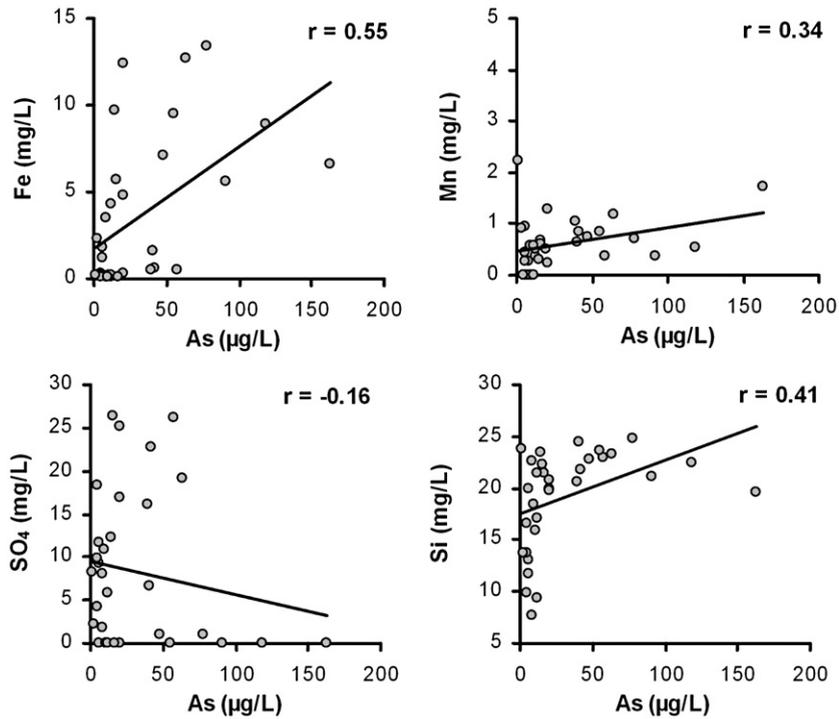


Fig. 8. Scatter plots showing statistical relationship among groundwater As, Fe, Mn, SO₄, and Si. Arsenic is positively correlated (see the Pearson's correlation) with Fe, Mn and Si in groundwater, but negatively correlated with SO₄ and such relationship is observed in many other As-contaminated areas in Bangladesh.

weighed and magnetically separated using a hand magnet and Frantz magnetometer for aid in mineral identification.

4. Results

4.1. Groundwater geochemistry

4.1.1. Major groundwater ions

Major ion compositions of groundwater samples from the study area are presented in Table 2. Chemical charge balance

between cation and anion in all groundwater samples ($n=85$) is reported as less than $\pm 5\%$ (Table 2). Spatial variations in the major ion concentrations in groundwater are considerable. Concentrations of all major ions in the Manikganj groundwater samples are shown in a box diagram (Fig. 4). The major cations in the sampled groundwater are Ca (32–43 mg/L) and Na (8–99 mg/L). Significant spatial variations are observed in the concentrations of Ca, Na, K, and Mg in groundwater. Concentrations of Ca in groundwater decrease with depth, whereas Na concentrations in

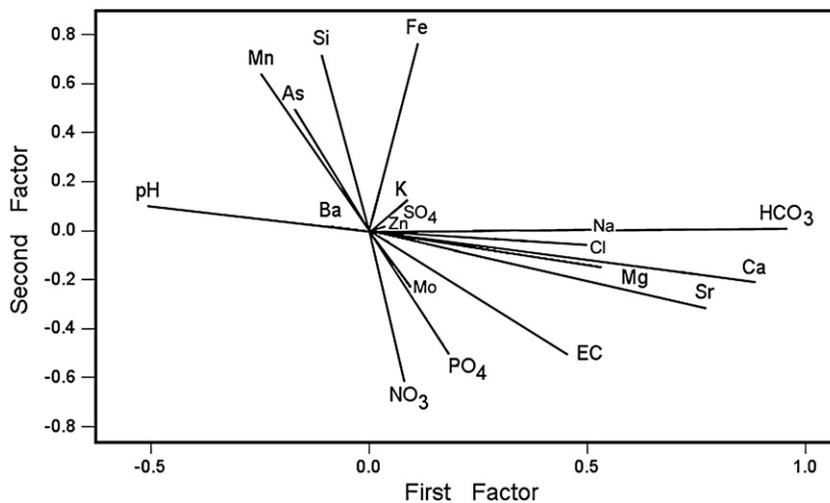


Fig. 9. Factor loading plots of Manikganj groundwater ions show that elevated concentrations of As, Fe, Mn, Si and high pH are closely associated with each other indicating origin from similar sources or by geochemical processes.

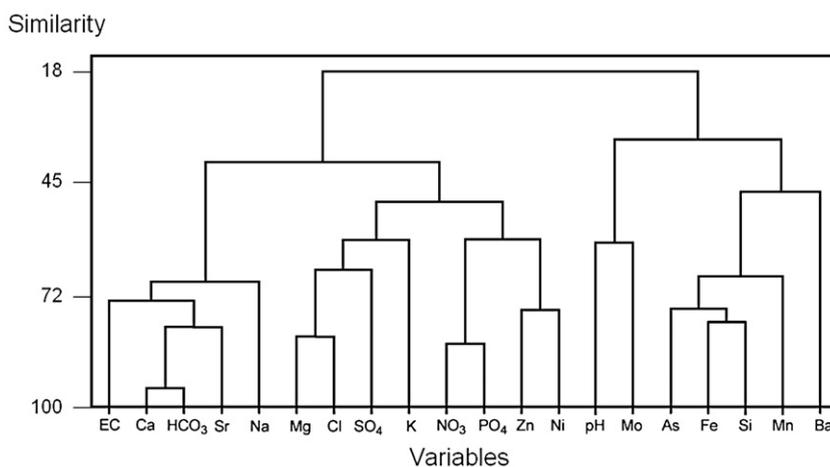


Fig. 10. Dendrogram is illustrating the major clusters in groundwater of the study area. Dissolved As, Fe, Si, Mn, and Ba form one of the major clusters in groundwater and suggest close geochemical association in aquifers.

groundwater increase with depth. Concentrations of Mg (13.3–46.1 mg/L) and K (0.67–40.0 mg/L) also decrease with increasing tubewell depths in Manikganj. Considerable spatial variations are observed in the concentrations of groundwater anions in Manikganj area. HCO_3 is the major anion of the groundwater in the study area, whereas Cl is the second dominant anion. Concentrations of HCO_3 (181.8–553.9 mg/L) are higher towards the southern part of Manikganj where groundwater SO_4 concentrations (0.01–75.7 mg/L) are also high.

Major ion compositions in Manikganj groundwaters plotted on a Piper diagram, indicate that the groundwater is mostly Ca– HCO_3 type (Fig. 5). Occurrence of high dissolved HCO_3 in groundwater in Manikganj is probably due to active biodegradation of organic matter mostly at shallow depths in aquifers (GRG and HG, 2002; Arafin, 2003; Turner, 2006). Fig. 5 also shows a distinct, but smaller difference in major chemical composition in Manikganj aquifers between shallow (<100 m) and deep groundwater (>100 m). Shallow groundwaters are high in Ca concentrations, but contain wide range of HCO_3 and SO_4 concentrations. Deeper groundwaters contain high concentrations of HCO_3 and Ca, but low concentration of SO_4 . Concentration of Ca varies slightly in deeper aquifers, but concentrations of HCO_3 range from 300 to 350 mg/L.

4.1.2. Arsenic and other trace elements

Concentrations of dissolved As, Fe, Mn, Si and other trace metals in groundwater of Manikganj area are given in Table 3. Considerable spatial and depth variations are observed in trace metals in groundwater of Manikganj study area. Several important trace metals including As, Fe, Mn, Sr, Zn, Ba and Si are shown in the Box and Whisker plots (Fig. 6) as these figures illustrate statistical variation in the concentrations of different groundwater constituents (Glynn and Plummer, 2005). Mean concentration of arsenic in sampled 85 tubewells in Manikganj is $\sim 33 \mu\text{g/L}$, even though the range is between 0.25 and 191 $\mu\text{g/L}$. The maximum arsenic level is 191 $\mu\text{g/L}$, which is almost 20 times higher than the WHO standard. The median arsenic concentration is

16.8 $\mu\text{g/L}$, and only 6 samples in 85 tubewells contain arsenic concentration of >100 $\mu\text{g/L}$. Considerable spatial distributions of groundwater arsenic are observed within Manikganj study area.

Fe is another important trace element commonly found at higher concentrations in Bangladesh groundwater (Table 3). In Manikganj, the average groundwater Fe concentration is 5.29 mg/L. The maximum Fe concentration is approximately 27 mg/L and the minimum is 0.001 mg/L. High Fe concentrations are found in tubewells where As concentrations are also higher than the average value of 33 $\mu\text{g/L}$. Considerable spatial distribution of Fe is seen in the study area (Fig. 7). Mn concentrations in the study area range from 0.001 mg/L to the maximum level of 4.16 mg/L with a mean value of 0.71 mg/L. In Manikganj, the spatial distribution of Mn in groundwater is also variable (Fig. 7). The groundwater has high concentrations of Si with an average value of approximately 19.4 mg/L. The maximum concentration of dissolved Si in Manikganj groundwater is 25 mg/L. The spatial distribution of Si in groundwater follows the distribution of arsenic in the northern part of the study area.

The concentration of dissolved Al in Manikganj groundwaters is very low (Table 3). The average Al concentration is approximately 0.01 mg/L. Concentration of Sr in groundwater varies from 0.06 mg/L to 0.68 mg/L with a mean value of 0.29 mg/L. High concentrations of Sr are found in tubewells that contain higher arsenic concentrations. Concentrations of dissolved Ba (0.013–0.80 mg/L), Zn (0.001–12.60 mg/L), Ni (0.03–2.4 $\mu\text{g/L}$), Mo (0.03–4.5 $\mu\text{g/L}$) and Pb (0.01–26.6 $\mu\text{g/L}$) are medium to low in groundwater of Manikganj area. Groundwaters with high As concentrations contain relatively high amount of Fe and Mn concentrations in most cases, but low in dissolved SO_4 concentrations. This relationship is consistent with other parts of Bangladesh where groundwater contains high concentration of As (BGS and DPHE, 2001).

4.1.3. Multivariate statistics on groundwater chemistry

Several multivariate statistical analyses are undertaken in this chapter to examine the relationship between arsenic and

Table 4

Concentrations of major, minor and trace elements in sediment samples (total digestion) from the MG and MN cores from the Manikganj study area

MG core sediment geochemistry												
Sample unit	Depth m	As mg/kg	Fe %	Mo mg/kg	Mn mg/kg	Ni mg/kg	Cu mg/kg	Pb mg/kg	Zn mg/kg	Co mg/kg	Sr mg/kg	Ca %
MG-002	2	7	4.29	0.5	659	65.0	60.0	32.2	97	20.4	34	0.72
MG-006	7	3.3	3.24	0.2	315	42.5	38.3	20.9	72	14.9	17	0.29
MG-008	9	5.5	3.55	0.3	434	48.8	44.0	21.6	77	16.7	26	0.69
MG-013	16	5.6	3.06	0.3	438	39.4	37.7	14.5	64	14.4	22	0.44
MG-017	20	0.6	0.82	0.1	103	10.2	2.4	5.7	19	3.7	6	0.12
MG-022	27	1.4	0.63	0.1	90	6.7	3.6	3.1	15	2.5	9	0.12
MG-030	36	0.8	0.61	0.1	82	6.4	2.3	3.0	18	2.4	8	0.14
MG-038	49	0.7	0.73	0.1	99	8.7	2.6	3.5	25	2.8	9	0.2
MG-041	67	0.5	0.94	0.1	122	11.3	3.9	4.2	20	4.0	7	0.12
MG-046	98	0.25	0.56	0.3	99	5.8	7.0	3.8	17	4.8	5	0.08
MG-049	116	0.6	2.13	0.5	127	23.2	12.0	12.9	52	10.7	15	0.26
MG-054	147	0.25	0.84	0.1	77	11.2	1.7	6.9	22	4.1	8	0.1
Sample unit	P %	La mg/kg	Cr mg/kg	Mg %	Ba mg/kg	Al %	Na %	K %	V mg/kg	Bi mg/kg	Cd mg/kg	U mg/kg
MG-002	0.05	18	64	1.18	146	2.74	0.04	0.33	66	1.1	0.10	4.1
MG-006	0.04	11	45	0.73	110	1.96	0.01	0.15	49	0.5	0.10	1.6
MG-008	0.04	15	46	0.97	122	2.34	0.02	0.29	54	0.8	0.10	3.0
MG-013	0.07	8	39	1.00	85	1.54	0.03	0.45	44	0.3	0.20	2.6
MG-017	0.02	5	10	0.23	20	0.47	0.01	0.12	12	0.1	0.05	0.5
MG-022	0.02	4	7	0.17	16	0.32	0.02	0.08	10	0.1	0.05	0.4
MG-030	0.03	4	7	0.16	13	0.3	0.02	0.09	11	0.05	0.05	0.4
MG-038	0.04	5	9	0.20	16	0.38	0.02	0.10	14	0.1	0.05	0.6
MG-041	0.02	4	11	0.27	22	0.49	0.02	0.14	16	0.1	0.05	0.3
MG-046	0.02	4	8	0.13	8	0.27	0.01	0.06	10	0.1	0.05	0.4
MG-049	0.02	22	31	0.45	47	1.41	0.01	0.28	24	0.3	0.05	1.0
MG-054	0.02	4	14	0.17	10	0.39	0.02	0.08	15	0.1	0.05	0.3
Sample unit	Depth m	As mg/kg	Fe %	Mo mg/kg	Mn mg/kg	Ni mg/kg	Cu mg/kg	Pb mg/kg	Zn mg/kg	Co mg/kg	Sr mg/kg	Ca %
MN-004	2	6.9	2.79	4.3	538	29.9	31.2	12.9	56	12.5	52	2.73
MN-007	4	8.8	3.20	0.5	749	35.9	38.9	20	85	13.2	40	0.66
MN-010	5	6.1	3.78	0.7	871	53.6	48.6	27.2	82	18.7	21	0.36
MN-014	7	2.6	3.13	0.3	560	44.0	29.4	11.3	75	16.1	10	0.31
MN-017	9	1.1	1.57	0.2	223	20.7	7.2	6.2	36	8.0	7	0.17
MN-023	13	0.9	1.10	0.1	128	12.9	4.0	3.6	22	4.5	5	0.13
MN-027	15	0.5	1.01	0.1	119	12.2	3.6	4.9	23	4.8	5	0.1
MN-032	18	0.6	1.17	0.4	202	11.5	7.9	2.7	21	5.3	9	0.13
MN-039	23	0.8	1.28	0.6	203	12.3	9.2	5.5	26	5.6	9	0.13
MN-047	27	0.5	0.81	0.5	111	9.7	2.2	4.3	16	3.8	4	0.08
MN-055	32	0.7	1.19	0.1	193	12.9	3.3	4.2	21	4.8	5	0.12
MN-061	36	0.5	1.00	0.1	129	11.8	3.3	3.5	19	3.7	8	0.23
MN-072	43	0.8	1.52	0.5	219	18.7	6.8	4.7	33	7.2	7	0.16
MN-077	46	0.6	1.11	0.1	162	14.0	3.6	5.3	24	5.0	5	0.13
MN-081	58	0.5	0.74	0.1	99	10.0	3.2	6.5	17	3.6	5	0.1
MN-084	67	0.6	0.87	0.1	128	10.9	5.8	8.3	17	4.2	7	0.12
MN-086	73	0.25	1.35	0.1	146	10.1	3.6	4.7	16	3.8	7	0.18
MN-092	92	0.25	0.75	0.5	105	9.4	3.3	4.4	17	3.5	5	0.11
MN-100	141	0.6	0.96	0.5	126	11.6	7.7	5.5	18	4.1	7	0.13
MN-102	152	0.25	1.46	0.1	130	19.6	6.0	4.4	32	7.5	10	0.17
Sample unit	P %	La mg/kg	Cr mg/kg	Mg %	Ba mg/kg	Al %	Na %	K %	V mg/kg	Bi mg/kg	Cd mg/kg	U mg/kg
MN-004	0.048	10	51	1.02	83	1.56	0.011	0.3	39	0.4	0.10	0.8
MN-007	0.349	10	40	0.74	173	1.74	0.012	0.38	44	0.5	0.10	3.8
MN-010	0.079	16	54	1.07	143	2.32	0.015	0.45	57	0.8	0.05	1.2
MN-014	0.049	6	39	1.02	86	1.66	0.017	0.64	47	0.3	0.10	2.1
MN-017	0.036	5	18	0.45	35	0.84	0.014	0.31	24	0.1	0.05	0.5
MN-023	0.039	4	13	0.29	23	0.54	0.009	0.15	17	0.1	0.05	0.6
MN-027	0.021	4	11	0.27	25	0.56	0.012	0.16	14	0.1	0.05	0.4
MN-032	0.028	4	11	0.22	24	0.41	0.011	0.13	15	0.05	0.05	0.4
MN-039	0.037	4	12	0.19	23	0.39	0.013	0.11	15	0.1	0.05	0.4
MN-047	0.017	4	10	0.21	20	0.43	0.007	0.12	11	0.1	0.05	0.3
MN-055	0.025	5	13	0.27	23	0.53	0.014	0.15	17	0.1	0.05	1.0

Table 4 (continued)

MG core sediment geochemistry												
Sample unit	P	La	Cr	Mg	Ba	Al	Na	K	V	Bi	Cd	U
	%	mg/kg	mg/kg	%	mg/kg	%	%	%	mg/kg	mg/kg	mg/kg	mg/kg
MN-061	0.062	5	12	0.28	20	0.49	0.012	0.12	16	0.1	0.05	0.5
MN-072	0.033	4	17	0.42	38	0.77	0.022	0.23	21	0.1	0.05	0.4
MN-077	0.024	4	12	0.3	27	0.58	0.009	0.17	16	0.1	0.05	0.5
MN-081	0.015	4	10	0.21	18	0.42	0.015	0.11	10	0.1	0.05	1.1
MN-084	0.022	4	11	0.21	17	0.41	0.015	0.1	12	0.1	0.05	0.7
MN-086	0.038	8	16	0.22	17	0.42	0.012	0.09	26	0.1	0.05	0.5
MN-092	0.022	4	9	0.22	16	0.42	0.007	0.1	11	0.1	0.05	0.5
MN-100	0.032	4	11	0.19	17	0.4	0.011	0.1	14	0.1	0.10	0.6
MN-102	0.031	5	18	0.34	16	0.69	0.007	0.16	22	0.1	0.05	0.3

other groundwater constituents (Glynn and Plummer, 2005). Pearson's correlation analysis was performed on multivariate properties of Manikganj groundwater on their log-transformed values. Results show that As is moderate to strongly correlated with Fe ($r=0.55$, $p=0.001$) and Si ($r=0.41$, $p=0.005$) in groundwaters (Fig. 8). Mn seems to be positively correlated ($r=0.34$, $p=0.038$) with groundwater As in Manikganj. Weak correlations also exist between pH and As ($r=0.30$, $p=0.5$) and Ba and As ($r=0.16$, $p=0.44$). As is negatively correlated with SO_4 ($r=-0.16$, $p=0.09$), electric conductivity ($r=-0.32$, $p=0.05$), HCO_3 ($r=-0.34$, $p=0.04$) and other ions in groundwater.

Factor analysis on log-transformed geochemical data reveals the principal elemental associations in groundwater (Stüben et al., 2003). In this study, factor analysis was limited to five factors that explain the significant portion of variance expressed by the data matrix (0.70% of communality). A varimax rotation algorithm was applied on the factors for easier interpretation of factor associations. Factor one explaining 24% of the variance has high loadings for groundwater Ca, Na, Mg, Cl, HCO_3 and EC and trace element Sr, and Ni. This factor represents the main dissolved load of groundwaters as a result of the interaction with minerals in aquifers and chemical weathering of catchment rocks. Factor two (18% of the total variance) includes As, Fe, Mn, K and Si in groundwater mainly suggesting a reducing condition (reductive dissolution of Fe- and/or Mn-oxyhydroxides) in aquifers and chemical dissolution of silica, phyllosilicate (biotite) and potassium minerals in groundwater. Strong to moderate statistical correlations among these variables were

also observed in correlation analysis earlier in this section. Factor three (12% of the variance) includes Cl, Mo, Ni, Zn and pH in groundwater. These trace metals (Mo, Ni, and Zn) in groundwater can be found due to reductive dissolution from Fe- and/or Mn-oxyhydroxide minerals (Bhattacharya et al., 2006). Normally, high Ni, Mo and other trace metals (As, Cr, Co, Pb, etc.) are adsorbed in Fe- and/or Mn-oxyhydroxides due to adsorption or ion-exchange during early diagenesis as coating. Higher Mo concentrations in groundwater are also expected at higher pH due to desorption from sediments (Goldberg et al., 1996). Factor four explaining approximately 10% of the total variance represents high factor loadings for Na, K, Ba, NO_3 , and PO_4 in groundwater as dissolved load of the water as a result of interaction with minerals in the aquifers. Factor five represents less than 10% of the variance and has high factor loading among As, Sr, and Mo. A moderate to strong positive correlation is frequently observed among As, Sr and Mo concentrations in groundwater samples from many As-contaminated aquifers in Bangladesh (BGS and DPHE, 2001). Sr in groundwater generally comes from the weathering of igneous rocks and carbonates. The factor score cross-plot between the first two factors is shown in Fig. 9. Groundwater As in Manikganj is closely associated with Fe, Mn, Si, Ba and pH, which is also observed in many other arsenic-contaminated groundwater aquifers in Bangladesh and West Bengal, India (BGS and DPHE, 2001; Dowling et al., 2002; Stüben et al., 2003; Hasan et al., 2007).

The log-transformed groundwater variables are used for cluster analysis with correlation coefficient as distance. Four clusters were formed after the final partitioning of variables

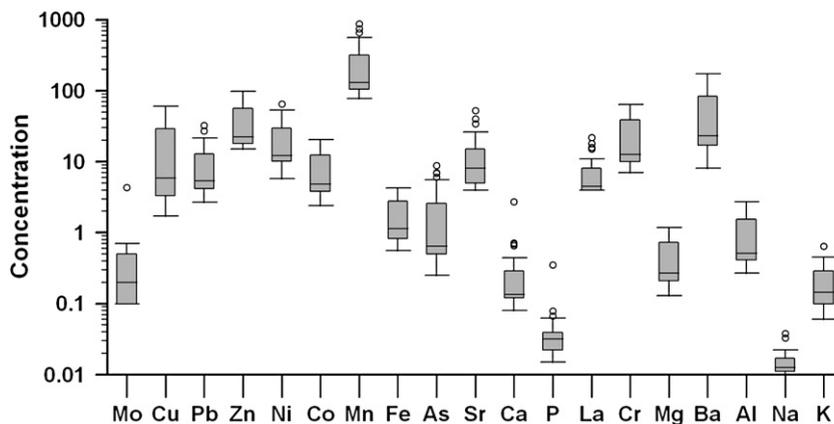


Fig. 11. Box plot showing variations of the chemical constituents' concentrations in the sediment samples in the study area. The inset diagram in Fig. 4 is referred as the key to this plot. Concentrations of Fe, Ca, P, Mg, Al, Na, and K are in % (10,000 mg/kg), but for all other constituents (including As), the concentrations are in mg/kg.

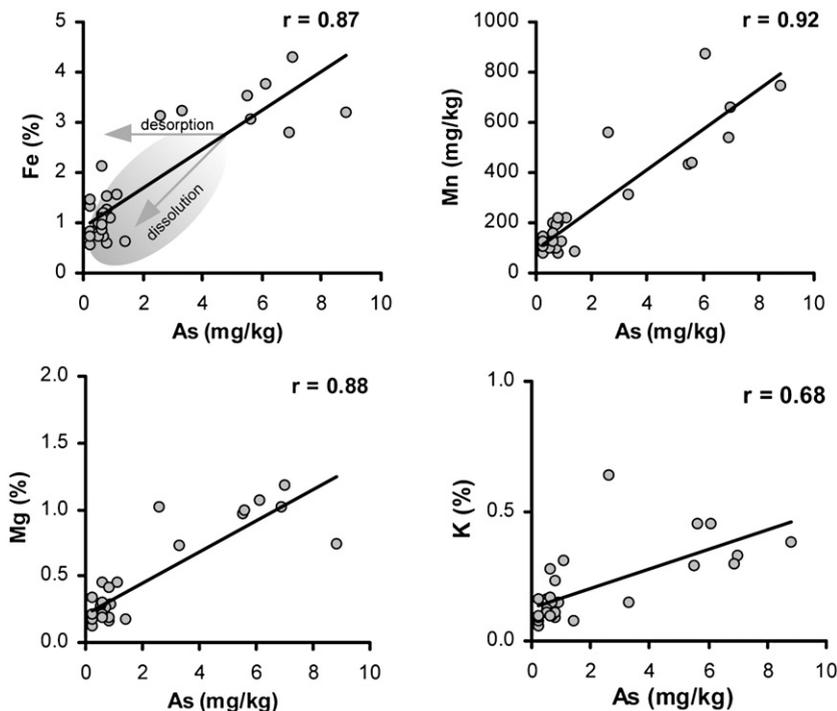


Fig. 12. Scatter plots showing statistical relationships (see the Pearson's correlation) among As, Fe, Mn, Mg, and K in sediment samples in the Manikganj MG and MN drilled cores. The theoretical model for the variations of Fe and As in the solid phase for the proposed mechanisms of the As release from Fe-oxides (or Fe-oxyhydroxides) (adopted from Gómez et al., 2006), suggests that the dissolution (reductive) processes were dominant over desorption as results of the analysis of both elements are concentrated in the grey area and alongside the regression line.

during the amalgamation process (i.e., linkage of clusters that are sufficiently similar) following the highest similarity levels among the variables. Clusters with their internal statistical similarity levels are represented by inverted tree structures or dendrograms (Fig. 10). Four major clusters were formed in groundwater constituents that indicate their internal relationship and sources of origin. Ca, Na, HCO_3 , Sr and EC form the first cluster in Manikganj groundwater as seen in the factor analysis. Ca, Na and HCO_3 represent the major ions in groundwater. Sr is strongly correlated with Ca, Na and EC in groundwater. Mg, Cl, K, SO_4 , Ni, NO_3 , PO_4 and Zn form the largest cluster in groundwater of Manikganj (Fig. 10). These elements that are predominantly influenced by chemical weathering of rocks and minerals represent the major groundwater chemistry. Cluster three is formed by pH and Mo in groundwaters. In groundwater, Mo occurs as an oxy-anion, and so its mobility should be similar to that of As (Smedley and Kinniburgh, 2002). Dissolved As, Fe, Mn, Si, and Ba in groundwater form cluster four that represents reductive dissolution of Fe- and/or Mn-oxyhydroxide minerals in aquifers as well as chemical break up of silicate and phyllosilicate (mainly biotite) minerals through the water-rock (sediment) interaction.

4.2. Sediment characteristics and geochemistry

4.2.1. Lithology and sediment characteristics

A total of 147 sediment samples from two drill-cores have been used for describing the general lithological characters of the sedimentary facies in the study area (Fig. 3). Based on

color, texture, grain-size, sorting and composition the sediments are classified into five sedimentary facies ("A" through "E"; Fig. 3). There are substantial variations in sediment grain-size, shape and colors within these sedimentary facies.

Most of the shallow sediments are composed of facies A (gray silt and silty clay). In the upper fining-upward sequence, gray silts and silty clay form thin-lamination within fine sands. At deeper depths (> 100 m), clay becomes sticky with very little silt. Facies A contributes less than 10% sediments in the study area. This facies is associated with floodplain or channel-fill deposits.

Facies B (gray fine sand) dominates the sedimentary deposits in Manikganj area. The intensity of gray varies from light to dark, and sometimes slight yellowish-gray. In many places at the vertical core profile this facies consists of muscovite and dark-colored heavy minerals. High amount of silt modifies this fine-grained sand facies at several depths. Some plant debris is found in this sedimentary facies but no other visible fossils were encountered in the study area. This facies was formed within the channel-bar and natural levee deposits.

Facies C (gray to yellow medium to coarse sand) forms the most common aquifer in the study area. These sediments were presumably deposited within the natural levees of a meandering channel towards the point-bar deposits during lateral migration of channel across the floodplain. Sediments at shallow depths are mainly grayish indicating reducing subsurface condition. This facies varies from gray to yellowish-gray towards the deeper cores indicating gradual change in geochemical environment in sequences. Minerals of this

facies are mostly quartz and feldspars with other dark-colored minerals. In most places this facies is moderate to well-sorted with no obvious sedimentary structures. Silts, fine sand to even coarse sands are also found within this sedimentary facies in several layers. High porosity and permeability make this facies one of the better aquifers in the study area.

The bottom part of the middle fining-upward sedimentary sequence is formed by facies D (gravel-rich clean sand) consists of gravel-rich medium to coarse sands. In both core sites, gravels of various compositions are found. At the MN site, the gravels are mixed with medium to coarse sands, which are moderate- to well-sorted. Sands are composed of clean quartz grains with feldspar and other heavy minerals. In MG core site, gravels are found within clay/silty clay layer, indicating channel lag to overbank deposits. Gravels content increases toward the bottom of the middle fining-upward sequence in the study area. The gravel-rich sandy deposits were probably formed in braided river-bed when sediment influx was high.

Typical yellowish-brown to bright orange-brown sands (facies E) are found in the lower fining-upward sequence in Manikganj indicating a highly oxidizing condition in sediments. Some yellowish-brown sands are also found in the middle fining-upward sequence, which are substantially different in texture and composition from the Dupi Tila sands. Gravels are found in these medium- to fine-grained sands (facies E) in some places. These sediments were probably deposited in migrating river channels in an active fluvial system.

4.2.2. Sediment geochemistry

Geochemical results from total digestion of sediment core samples are given in Table 4, which shows variations in concentrations of 36 different elements from shallow to deep aquifers in Manikganj. Fig. 11 shows the concentrations of the major chemical constituents in core sediments in the study area. Arsenic concentrations in sediments range from 0.3 mg/kg to about 8.8 mg/kg, with an average concentration of approximately 2.2 mg/kg and 1.7 mg/kg in MG core and MN core respectively. Ranges of arsenic concentrations are approximately 3.7 mg/kg and 7.7 mg/kg within 15 m (~50 ft) and 9 m (~30 ft) depth at MG and MN core samples respectively. Mean concentrations of Fe in sediments are 1.78% and 1.54% in wells MG and MN respectively. Maximum concentrations of Fe are 4.26% in core MG and 3.78% in core MN in Manikganj. The mean concentrations of Mn are approximately 220 mg/kg and 257 mg/kg in core MG and MN respectively. Arsenic concentration in sediments is found to be very low (<1.5 mg/kg) at any depth below 50 ft from the surface. Concentrations of other trace elements in sediments (e.g., Ni, Cu, Pb, Zn, Sr, Co, La, Bi, V etc.) are also higher in the high arsenic zones. Multivariate statistical correlation analysis was performed on elemental concentrations in sediments to examine the relationships between arsenic and other elements. Results suggest that As in Manikganj sediments is strongly correlated with Fe ($r=0.87$), Mn ($r=0.92$), Mg ($r=0.88$), K ($r=0.68$), Ca ($r=0.90$), P ($r=0.89$) and other trace metals such as Zn ($r=0.87$), Ni ($r=0.84$), Co ($r=0.85$), Cr

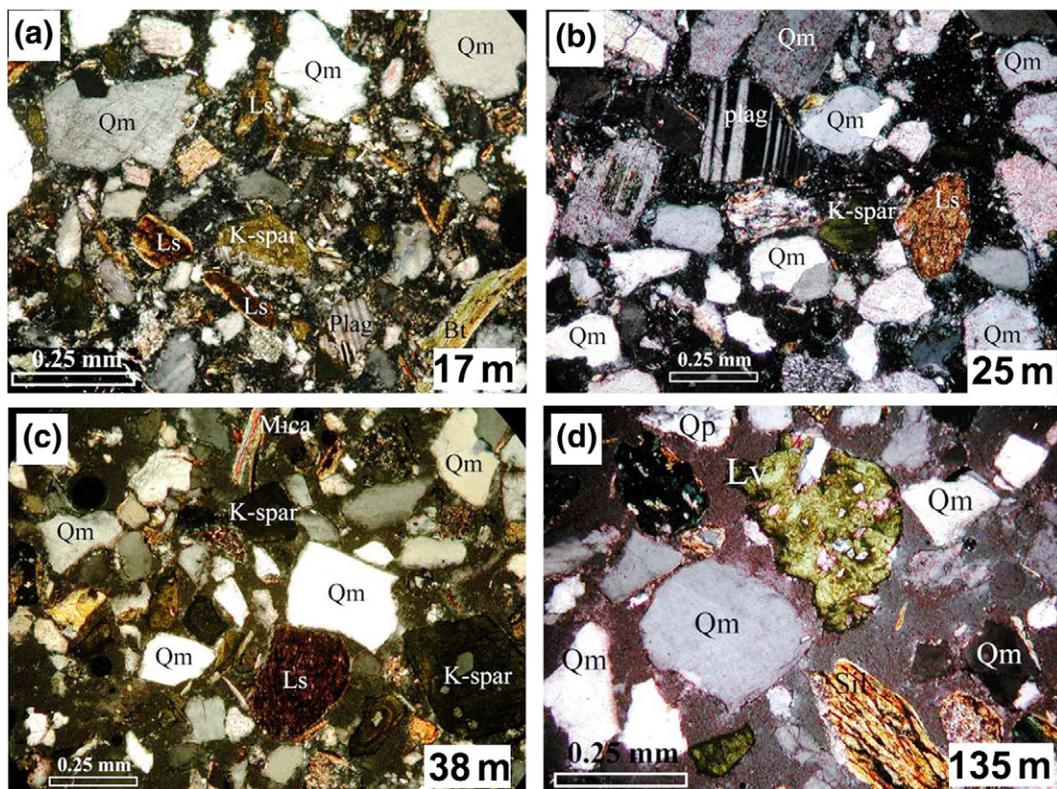


Fig. 13. Representative photomicrographs of sands from Manikganj study area. The sample depths are given on the plates. Keys: Qm – Monocrystalline Quartz; Qp – Polycrystalline Quartz; K-spar – Potassium feldspar; Plag. – Plagioclase feldspar; Ls – Sedimentary lithic; Lv – Volcanic (plutonic) lithic; Lm – Meta-morphic lithic; Gt – Garnet; Sil – Sillimanite; Bt – Biotite.

($r=0.87$), Cu ($r=0.89$), La ($r=0.73$) and Sr ($r=0.89$). In these correlations, the p value is always <0.001 , which suggests that the correlations are significant. Fig. 12 shows representative scatter plots among As and other major constituents in sediments. The scatter plot between Fe and As indicates that mineral dissolution (reductive) is involved in mobilization of Fe and As in groundwater (as suggested by Gómez et al., 2006). However, presence of higher concentrations of As and Fe in sediments suggests that some desorption process may result in such pattern (Fig. 12), which indicates that high As was released from samples (rich in detrital biotite) originally containing high Fe concentration.

4.2.3. Petrography, mineralogy and sediment provenance

The mineralogy of sediments both in shallow and deep aquifers are mainly quartz (60–70%), feldspar (20–25%), both potassium and plagioclase, lithic fragments (10–15%) and micas (2–5%) (Fig. 13). Quartz is both monocrystalline and

polycrystalline varieties with variable undulosity. Feldspars are mostly potassium type (up to 20% of the bulk sediment composition) at shallow depths with some plagioclase feldspar mainly albite and Ca-rich varieties. Perthitic and myrmekitic intergrowth structures have been observed. Lithic fragments are mainly sedimentary (e.g., shale, mudstone, argillite and siltstone) with some low- to medium-grade metamorphic lithic grains at shallow depths, but medium- to high-grade metamorphic lithic grains with less sedimentary lithic grains are abundant in the sediments at deeper depths. Plutonic igneous fragments are also abundant throughout the Quaternary sediments in Manikganj (Fig. 13). Based on petrographic analysis, the sands in the shallow aquifers are classified as arkose to subarkose type.

Point-count data were plotted in various ternary diagrams in order to suggest provenance information. The samples from the upper Cenozoic sediments (Holocene alluvium and Pliocene–Pleistocene Dupi Tila sand) fall within the border

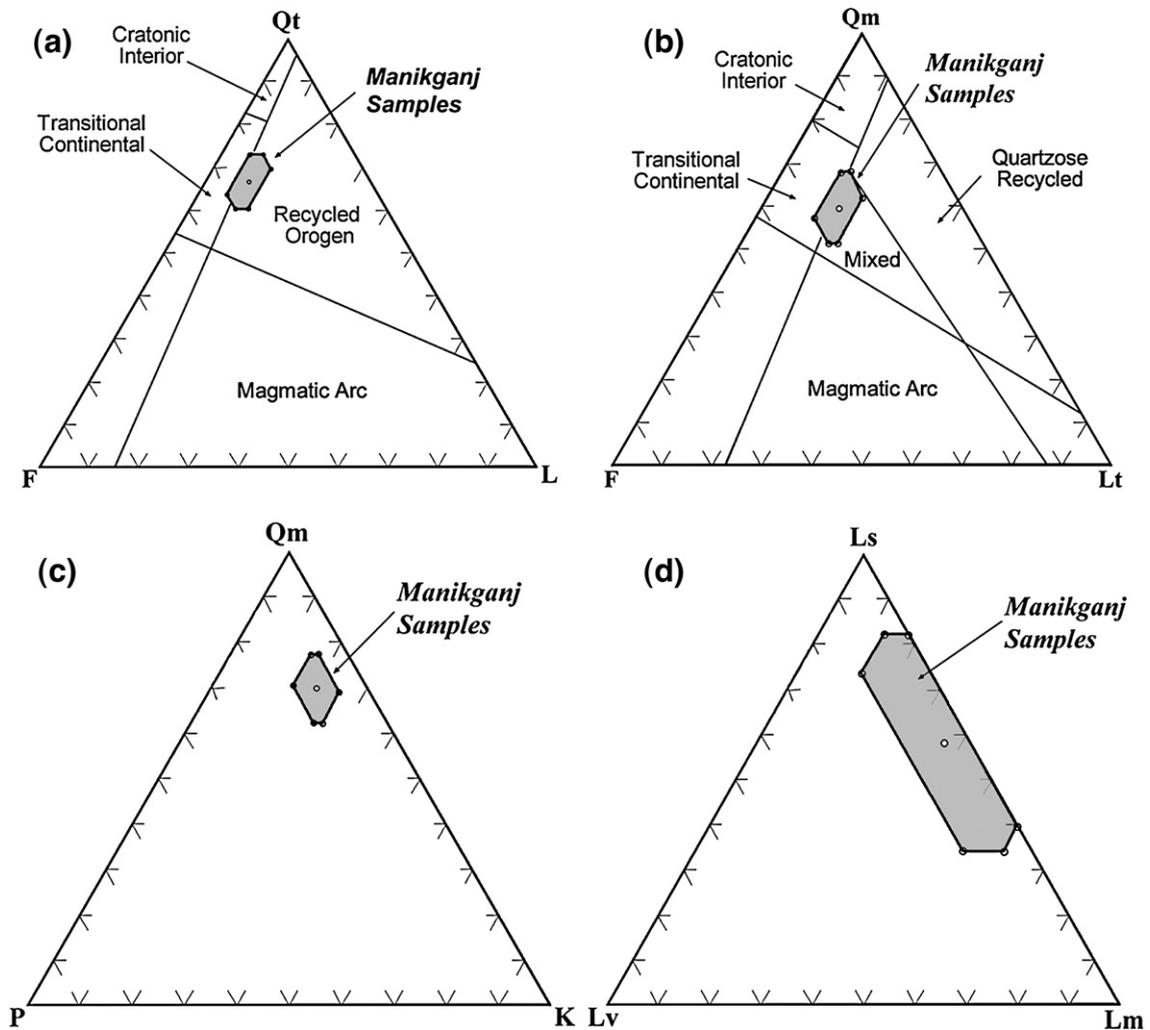


Fig. 14. Ternary diagrams showing overall provenance modes in (QtFL and QmFLt plots), light microcrystalline component (QmPK), and lithic-grain component (LsLvLm) of sand composition. (a) QtFL plot shows quartz- and feldspar-rich composition lie within the border between “recycled orogen” and “transitional continental” provenances; (b) QmFLt plot shows that sands are derived from mixed tectonic provenance; however, contributions from “transitional continental” are also discernible; (c) Quaternary sands from Manikganj area are dominated by potassium feldspars; (d) Lithic fragments are mainly sedimentary (e.g., mud, shale, silty sand) and low-grade metamorphic rocks (slate, phyllite, schist and gneiss). Provenance fields in (a) and (b) are taken from Dickinson, (1985).

between “recycled orogen” and “transitional continental” fields (Fig. 14). These suggested fields are slightly different from the provenance field (completely “recycled orogen”) recommended for the Dupi Tila Sandstone at a comprehensive study on provenance and tectonic history of the Bengal Basin by Uddin and Lundberg (1998a).

Average concentration of heavy minerals is approximately 4.36 (wt.%) with the maximum concentration of 10.28 (wt.%) and the minimum of 2.44 (wt.%) in the Manikganj sediments (Shamsudduha, 2007). Highly magnetic minerals are mainly magnetite, ilmenite, and hematite that contribute approximately 5% of the heavy minerals. The magnetic minerals are fairly homogenous with minor Ti content indicating low Ti-magnetite that appears to be plutonic in origin. Ilmenite is the second largest variety of magnetic minerals, which contains very high amount of Ti and Fe. Moderate magnetic minerals are garnet, biotite, chlorite, ilmenite, and some amphiboles containing magnetite inclusions. This is the largest (~50%) group of heavy minerals observed in Manikganj core samples. The second (37%) group in which the dominant heavy mineral assemblages are amphibole, epidote, allanite, biotite, chlorite, staurolite, zoisite, pyroxene, and authigenic siderite. Sillimanite, kyanite, zircon, sphene, apatite, and some rutile are found in the poorly magnetic fraction, which contributes approximately 8% of heavy minerals in the core samples.

Although, some detrital pyrite grains are identified in shallow sediments, but no authigenic pyrite has been found in sediments from the Manikganj study area (Shamsudduha, 2007). Tourmaline, monazite, corundum and olivine are found in some samples. The concentrations of heavy minerals in the Quaternary sediments are higher than Cenozoic sediments in the Bengal Basin (Uddin and Lundberg, 1998b).

5. Discussion

5.1. Hydrogeochemistry and groundwater arsenic

The chemistry of groundwater in Manikganj area is similar to other arsenic-affected areas in Bangladesh. As is closely associated with Fe, Mn, Si and Ba in groundwater and sediments in the Manikganj study area. These elements in groundwater typically shows a moderate to strong correlation in other parts of Bangladesh where groundwater arsenic concentration is high (BGS and DPHE, 2001; Ahmed et al., 2004; Zheng et al., 2004). The breakdown of organic matter by bacteria influences the groundwater As concentration by serving as an electron donor for the reductive dissolution of Fe- and Mn-oxyhydroxides in aquifer sediments. Statistical correlation between groundwater As and SO_4 in the study area is negative, which is the common relationship in other

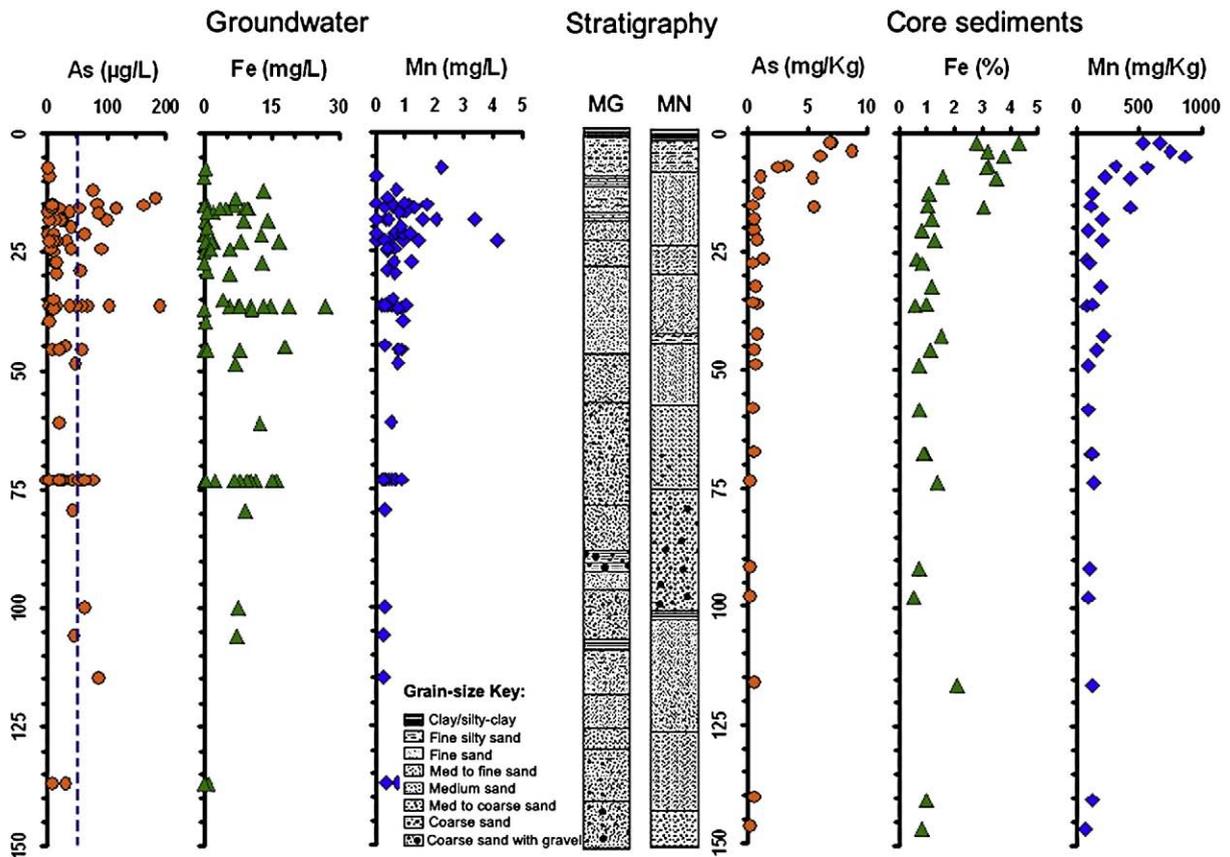


Fig. 15. Diagram shows the depth-wise variations in the concentrations of As, Fe and Mn both in groundwaters and sediments in the study area. Stratigraphic columns (MG and MN cores) are also shown with depths (m). Concentrations of these three statistically correlated chemical constituents are higher within the first 50 m below the groundwater surface.

high As areas in Bangladesh and West Bengal (Stüben et al., 2003; Ahmed et al., 2004; Zheng et al., 2005). A recent As-bioremediation research in our study area (Saunders et al., in press) has reported that shallow groundwater (depth <100 m) is limited to low SO₄ concentration and the dominant chemical processes are Fe- or Mn-reduction or combination of both. Their research also showed that stimulation of sulphate-reducing bacteria by adding molasses (source of organic carbon) and Epsom salt (MgSO₄·7H₂O) triggered the biogenic SO₄-reduction and decreased As concentration to <10 µg/L from a background level of 200 µg/L. Both factor and cluster analyses results show that As, Fe and Mn in groundwater and sediments are closely associated in As-affected shallow aquifers in Manikganj. Multivariate statistical results suggest that As in sediments is strongly correlated with Fe, Mn, Ca, P and other trace metals (e.g., Zn, Ni, Co, Cr, Cu, La and Sr) in sediments. Fig. 15 shows depth-wise variations in the concentrations of As, Fe and Mn both in groundwater and sediments. Higher concentrations of these three elements in aquifer sediments and water suggest that these constituents were derived from similar geochemical processes and most likely from similar mineralogical sources. It has been widely accepted that Fe and Mn are dissolved into groundwater through the reductive dissolution process from Fe- and Mn-oxhydroxides and this dissolution process releases As, ferrous Fe and Mn into groundwater (Nickson et al., 2000; BGS and DPHE, 2001; Zheng et al., 2004; Saunders et al., 2005). Besides, high Si is often found with high As in groundwater that might be derived from chemical weathering (and dissolution) of phyllosilicates minerals (e.g., biotite) in aquifers and can release high As in groundwater (Sengupta et al., 2004).

5.2. Quaternary sedimentation and climatic effects on arsenic distributions

Sedimentation patterns and deposits during the Quaternary period are different from the Tertiary and older geologic time in the Bengal Basin (Uddin and Lundberg, 1999; Goodbred and Kuehl, 2000). High groundwater arsenic is well-linked with the Quaternary deposits and no arsenic has been reported from the older sediments in Bengal Basin (Uddin and Abdullah, 2003).

In Manikganj, the As-free deeper aquifers (>100 m) are composed of highly oxidized and weathered Dupi Tila sands, which was probably deposited during Late Pliocene to Early Pleistocene (Reimann, 1993). Yellowish-brown to orange-brown sediments are mostly composed of quartz, feldspar, mica and some heavy minerals. The Dupi Tila sands contain less organic matter than the overlying As-rich Holocene sediments that are mainly fine-grained, gray to dark-gray in color. Percentage of heavy minerals in the Holocene sediments that form the shallow aquifers (<100 m) in Manikganj is higher than the older deposits. In the Manikganj stratigraphy, a reddish-brown clay layer separating the Dupi Tila sands from the overlying Holocene deposits is highly oxidized and sticky, and can be correlated with the Pleistocene Madhupur Clay (Shamsudduha and Uddin, 2007). However, this clay layer was greatly eroded during the last glacial maximum and sea-level lowstand in the early Holocene time (Goodbred and Kuehl, 2000). In Manikganj area, the Holocene

sedimentation started with gravel-rich medium to coarse sands recorded at approximately 100 m below surface (Fig. 3).

Sedimentation rate was very high and rapid as indicated by the presence of partly broken prismatic quartz crystals and very unstable minerals like olivine in the core samples from Manikganj. Generally, in the older sediments in the Bengal Basin, the presence of such chemically unstable minerals (olivine) was very rare (Uddin and Lundberg, 1998b). Significant sedimentation started in the early Holocene time with increased precipitation in warm and humid climatic conditions in the Himalayan region (Goodbred and Kuehl, 2000). Two distinct fining-upward sedimentary sequences are recorded in the Quaternary stratigraphic column probably indicating episodic major channel shifting in the study area. The bottom fining-up sequence, which is relatively coarser and contains low arsenic, was probably deposited by a braided river during the Pliocene–Pleistocene time. Sedimentation during the Middle to Late Holocene time in the study area was probably contributed by a meander river system relatively smaller than the present-day Brahmaputra river, which was apparently flowing through its easterly course into the Sylhet trough (Goodbred and Kuehl, 2000). Sediments in the upper fining-up sequence are predominantly fine-grained with clay and peat in several depths indicating the limit of the Holocene sea-level highstand when extensive mangrove forests and swamplands were developed in the Bengal Basin (Umitsu, 1987; Goodbred and Kuehl, 2000). This upper fining-up sequence forms most of the high arsenic-contaminated aquifers in the study area as well as in other parts of Bangladesh.

5.3. Lithology, mineralogy and provenance of As-rich aquifer sediments

General lithological and mineralogical compositions of the sediments both in arsenic-affected and arsenic-free aquifers in the study area are very similar. However, fine to coarse sediment ratio is smaller in the arsenic-free deeper (>100 m) aquifers which is similar to other areas in the country (Ahmed et al., 2004). These aquifer sediments vary from gray at the shallow depths to yellowish-brown at deeper depths indicating a reducing to oxidizing geochemical environments. Studies have shown that high As concentrations are associated with gray to dark-gray sediments, whereas yellowish-brown aquifer sediments host low groundwater arsenic (BGS and DPHE, 2001; Horneman et al., 2004).

Provenance analysis revealed that these Quaternary sediments were derived mostly from igneous and metamorphic terranes located in the Indian craton, Himalayan Mountains, Shillong Plateau and Indo-Burman ranges. Provenance field analysis shows that the sands from Quaternary deposits fall within the border between “recycled orogen” and “transitional continental.” The Ganges, Brahmaputra and Meghna rivers have distinctive sedimentary characteristics which result from geologically distinct source areas and catchment geomorphology. The Brahmaputra drains the Tibetan Plateau of China and Shillong Plateau of Assam and is dominated by upland tributaries originating from the Himalayan mountain ranges (Heroy et al., 2003). This river flows through rock types including Precambrian metamorphic (high-grade schists, gneisses, quartzite, and metamorphosed limestones), felsic

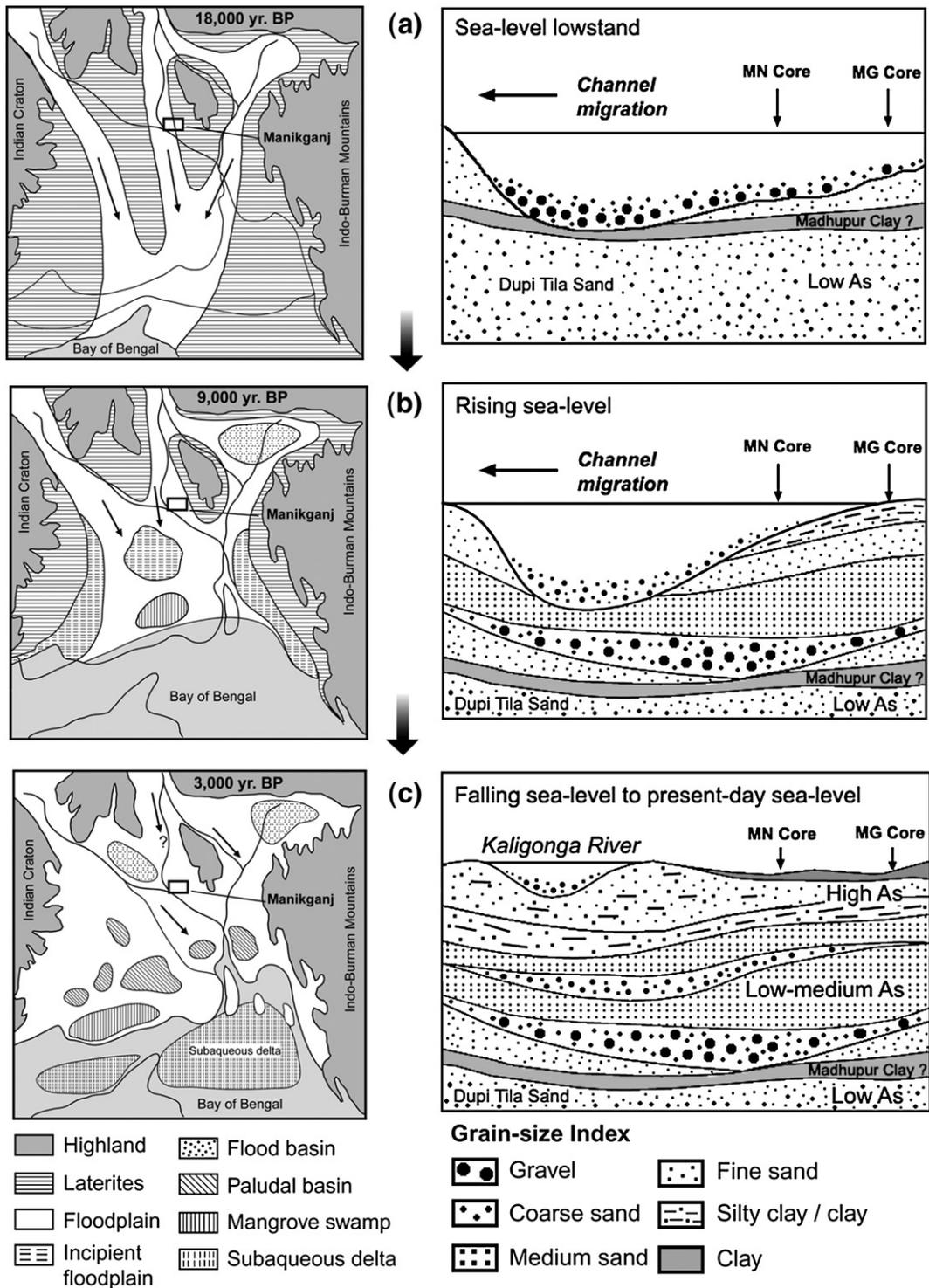


Fig. 16. Paleogeographic reconstruction of the Bengal Basin through the Quaternary period (after Goodbred and Kuehl, 2000) and postulated sedimentation patterns in Manikganj. (a) Low sea-level condition and sedimentation only within the incised river valleys; (b) Rapid sea-level rise and infilling of channel beds and adjoining floodplains; (c) Sea-level fall and appearance of the modern delta, and fine-grained deposits within numerous extensive peat basins and mangrove swamps that are highly As-contaminated.

igneous intrusive, and Paleozoic–Mesozoic sandstones and limestones (Huizing, 1971; Heroy et al., 2003). The Ganges travels through similar rock types, but unlike the Brahmapu-

tra, it flows through a vast Precambrian Indian craton. The Ganges is also fed by many tributaries draining the Mesozoic and Tertiary mafic effusives, Rajmahal traps, and Gondwana

basins in the eastern India that contain bituminous coals. The Meghna drains most of the western side of the Indo-Burman ranges through Tertiary sandstones, shale and limestones and igneous intrusions. Heavy mineral assemblages in these Quaternary sediments in the study area suggest a mixed source of origin where contributions from high- to medium-grade metamorphic rocks, plutonic igneous rocks, and pre-existing sedimentary rocks are significant. Results from our study show that there is no primary sources of As in sediments and therefore in groundwater. Mineralogical studies show that aquifers that are rich in dissolved As concentrations are also rich in Fe- and Mn-oxyhydroxides, phyllosilicate minerals (mainly biotite), iron-oxides (magnetite), and amphiboles. Similar mineralogical associations were reported by Acharyya and Shah (2007) from their study in the Gangetic alluvial plains in the West Bengal, India. The weather and erosion rates within the Himalayan Mountains are also variable. Guillot and Charlet (2007) have reported that around 25% sediments drain into the Bay of Bengal through the Siang and Brahmaputra rivers; the Ganges contributes about 40% sediment supplies and the rest comes from other Himalayan drainage systems. The provenance results from our study suggest that a major portion of the Quaternary sediments was derived from the orogenic belts, including the Miocene–Pliocene Siwalik sediments of the southern Himalayas. Recent studies in Terai plain of Nepal and in Pakistan proposed that the occurrence of high As in groundwaters is related to high erosion of Siwalik sediments during the Holocene time (Shrestha et al., 2003; Nickson et al., 2005; Williams, 2005). However, mineralogical assemblages in the Manikganj study area also suggest that a portion of the sediments was derived from the weathered ophiolites in the suture zone in the Himalayas and Indo-Burman Mountains (Uddin and Lundberg, 1998b), which contain peridotite and other olivine and amphibole rich mafic and ultra-mafic rocks. A recent study (Guillot and Charlet, 2007) has proposed that Indus-Tsangpo suture zone dominated by arc-related rocks and particularly serpentinites enriched in arsenic could be the primary source of arsenic in groundwater in the Bengal Basin.

5.4. Geomorphic evolution through Quaternary period and arsenic distribution

Occurrences of high groundwater arsenic are significantly linked with the geology–geomorphology of development of deltas and alluvial deposits in the Bengal Basin throughout the Quaternary period (Ahmed et al., 2004; Ravenscroft et al., 2005; Shamsudduha and Uddin, 2007). The Quaternary sedimentation and river dynamics were controlled by the global climatic changes, uplift of the Himalayas, and tectonic subsidence in the Bengal Basin (Umitsu, 1993; Goodbred and Kuehl, 2000). During the sea-level lowstand at 18,000 yr BP, much of the GBM system was occupied with deeply incised channels within a series of terraces that are covered by thick Holocene deposits (Ravenscroft et al., 2005). Much of the Bengal Basin comprised incised alluvial valleys and exposed lateritic uplands during the last sea-level lowstand (Fig. 16). The pre-existing sediments (Pliocene–Pleistocene Dupi Tila sands) were exposed and deeply oxidized. Steeper hydraulic

gradients allowed sufficient flushing of the Dupi Tila aquifers and recrystallization of iron-oxyhydroxides in dry climatic conditions (Ravenscroft et al., 2005). Sedimentation started with the climatic warming during the Late Pleistocene to Early Holocene, when voluminous sediment poured into the basin and filled the deep river valleys with coarser deposits (Fig. 16; Goodbred and Kuehl, 2000). During the rising sea-level conditions (11,000–6000 yr BP) relatively fine-grained sediments started depositing over the coarser sediments that deposited earlier and the sediment depositor migrated toward onshore (Goodbred and Kuehl, 2000). In Manikganj area, the Brahmaputra was probably depositing medium to fine-grained sediments forming fining-upward sedimentary sequence as recorded in both core samples (Fig. 3). Increased rainfall and humid climatic conditions favored the formation of mangrove swamps and peat basins behind the advancing delta fronts where degradation of abundant organic matter started (Ravenscroft et al., 2005). These sediments form the lower shallow aquifers in Manikganj area that contain low to medium concentrations of arsenic (0–50 µg/L) in groundwater. The delta progradation started when the rate of sea-level rise slowed and the maximum transgression was reached in the western Bengal Basin (Umitsu, 1993; Goodbred and Kuehl, 2000). The Brahmaputra river switched to its eastern course and drained into the Sylhet trough from ~7500 to 6000 yr BP. In the western side of the Madhupur Tract, sedimentation was slower and mostly fine-grained sediments were deposited in the wide floodplains as noticed in core samples from Manikganj. Subdued topography resulted in sluggish groundwater movement with little flushing that favored accumulation of organic matter, finer sediments and Fe-oxyhydroxides in the floodplains and natural levees, while sediments within the riverbeds were mostly coarse-grained (Ravenscroft et al., 2005; Shamsudduha and Uddin, 2007). Between 5000 and 6000 yr BP, the Brahmaputra probably switched back to its western course (Goodbred and Kuehl, 2000). Extensive paludal basin and mangrove swamps in the Bengal Basin were developed on the previously flooded coastal platform as the sea-level started dropping slowly and eventually stood at its modern stand. Mostly fine-grained sands and silts with clay were deposited in Manikganj area that formed the upper shallow alluvial aquifers, which contain high arsenic concentration. These sedimentary facies, which were formed by meandering rivers are characterized by medium to fine channel sands, overbank mud, and some peats that are noticed in Manikganj core sediments. Accumulation and degradation of organic matter and reducing conditions triggered arsenic release from Fe-oxyhydroxides due to microbially mediated reductive dissolution process at shallow depths. In Manikganj cores, the relative abundance of arsenic in sediments is higher in the MG core than the MN core. At shallow depths, the MG core samples are dominated by fine sands, silts and clay than the MN core samples.

6. Conclusions

This study has integrated the results of groundwater geochemistry with sediment characteristics and chemistry, petrography and mineralogy of aquifer sediments from the

Manikganj town in the central Bangladesh. Results show that the elevated arsenic concentrations in the shallow (<100 m) alluvial aquifers are associated with gray, fine-grained over-bank and floodplain deposits enriched with organic matter and peat. Petrographical and mineralogical results show that the sands in As-contaminated shallow aquifers are mainly arkose to sub-arkose and composed of high percentage of quartz, potassium feldspar, iron-oxides, biotite, magnetite, and amphiboles. Geochemical results show that shallow sediments that are rich in iron-oxides minerals contain As concentrations of as high as 8.8 mg/kg. There is no As-enriched primary mineral found in the aquifer sediments in the study area. High As levels in sediments are suspected to be associated with Fe-oxyhydroxides, detrital magnetite, apatite, biotite and some amphibole minerals in shallow aquifers. Microbially-mediated reductive dissolution of amorphous/colloids and crystalline Fe-oxyhydroxides (e.g., goethite) is thought to be the principal mechanism for the occurrence of high arsenic in alluvial aquifers in Manikganj as well as other similar fluvial environments in Bangladesh. Small-scale spatial variations in arsenic distribution might be controlled by the local variation in redox conditions (Mn/Fe reduction vs. SO₄ reduction) and distribution of sedimentary facies and minerals in the alluvial aquifers. The Siwalik sediments and As-rich ophiolites in the Himalayas and Indo-Burman Mountains are most likely the major primary sources of As in groundwater in the Bengal Basin.

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