Quaternary shoreline shifting and hydrogeologic influence on the distribution of groundwater arsenic in aquifers of the Bengal Basin

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Abstract

Naturally occurring high arsenic concentration in groundwater of most alluvial aquifers in the Bengal Basin has been causing serious health problems in millions of people. Elevated dissolved arsenic concentrations are mostly confined within a shallow depth (<150 m) of the Middle Holocene aquifers of the GBM delta and the rapidly subsided Sylhet trough. Arsenic-rich zones in the Bengal Basin are located in the south-central parts of Bangladesh and northeastern parts of West Bengal, India, bounded by Chittagong Hills in the east and the Indian Craton to the west. Holocene sea level rise and development of reducing conditions at organic-rich swampy lands are directly linked to epicenters of arsenic distributions. Surface elevation and topographic slope seem to control the distribution of arsenic because higher levels of dissolved arsenic occur mainly within the present-day topographically low areas. Delta lobes that have experienced tidal influx in the recent past do not appear to have high arsenic concentrations in groundwaters. Groundwater quality data suggest that the sulfate-reducing condition in the coastal aquifers may limit the dissolved arsenic and iron concentrations in aquifers.

Keywords: Arsenic; Groundwater; Hydrogeology; Sea level change; Paleoshore; Bengal Basin

1. Introduction

Naturally occurring elevated arsenic (As) in groundwater of the Bengal Basin, including Bangladesh and West Bengal of India has been recognized as the worst case of groundwater contamination in the world. Millions of tube-wells were installed in the Ganges–Brahmaputra–Meghna (GBM) delta complex in the last four decades to provide pathogen-free water for domestic and irrigation purposes in Bangladesh and West Bengal (Smith et al., 2000; BGS and DPHE, 2001). The major switch from polluted surface water to groundwater helped people avoid waterborne diseases, but detection of elevated dissolved arsenic in groundwater has panicked the people of Bangladesh and West Bengal, India (Nickson et al., 2000; BGS and DPHE, 2001). The first reported case of high arsenic in groundwater from the West Bengal of eastern India was recorded in 1978 (Acharyya et al., 2000). In 1993, the Department of Public Health Engineering (DPHE) first reported the existence of arsenic poisoning in the groundwater of Bangladesh in an area bordering West Bengal, but it was not until 1995 that the extensive occurrence of high arsenic was widely known (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, found that nearly 35 million people were drinking groundwater containing As with a concentration of more than 50 μg L⁻¹ (Bangladesh standard), and about 57 million people consume water that exceeds 10 μg L⁻¹ As (World Health Organization standard), mostly extracted from alluvial aquifers located within 10–50 m of the ground surface (BGS and DPHE, 2001). In West Bengal, about 5 million people in nine districts in the southern deltaic region are found to be badly affected by arsenic poisoning in groundwater (Acharyya et al., 2000; SOES, 2006).
Arsenic contamination of natural origin in groundwater has also been reported in many other parts of the world, including Argentina, Australia, China, Chile, Pakistan, Taiwan, Thailand, Mexico, Vietnam, and many parts of the United States (Smedley and Kinniburgh, 2002; Nickson et al., 2005; Liu et al., 2006). However, the human health effects of groundwater arsenic in the Bengal Basin are the most widespread. The occurrence, origin, and mobility of arsenic in groundwater vary among the contaminated sites around the world (Ravenscroft et al., 2001). The mode of occurrence and mobility of arsenic in sedimentary aquifers is 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 the Bengal Basin, the occurrence of arsenic and its mobilization is associated with geochemically reducing subsurface environment. Several recent studies agree that biogenic reductive dissolution of Fe-oxyhydroxides is the primary release mechanism that puts arsenic into groundwater in Bengal Basin alluvial aquifers (Bhattacharya et al., 1997; Nickson et al., 1998; Zheng et al., 2004). A similar mechanism was proposed to explain arsenic contamination in Taiwan, Vietnam, and parts of the United States (Saunders et al., 2005; Liu et al., 2006). 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 largescale irrigation pumping. Saunders et al. (2005) tried 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 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 the low temperatures during erosion (McArthur et al., 2004). Close connection between groundwater arsenic and presence of glacial deposits was observed in many places in North America and Europe (Saunders et al., 2005).

Spatial distribution of high contents of As in groundwater contained in alluvial aquifers in Bangladesh and West Bengal is not random, rather it is controlled by regional hydrogeologic setting and geologic–geomorphic units of the country (Ahmed et al., 2004; Shamsudduha et al., 2006a). However, arsenic concentrations at shallow depths within the same aquifer and at similar depths are unpredictable (van Geen et al., 2003; Shamsudduha, 2004). High concentrations of groundwater arsenic, and the highest probability of exceeding Bangladesh standard of 50 μg L⁻¹, most often occur in tubewells screened within 50 m of the ground surface (Ravenscroft et al., 2001). Similar conditions exist in the western part of Bengal Basin, where the highest arsenic concentrations both in groundwater and aquifer sediments are found within a few meters to about 50 m below the ground surface (Pal et al., 2002).

This study examines the spatial and depth distribution of arsenic contamination in alluvial aquifers and relates this distribution to Quaternary sea level, hydrogeology and surface elevation in the Bengal Basin. Groundwater arsenic distributions in aquifers combining both Bangladesh and West Bengal are mapped using GIS techniques to understand the spatial distribution on a basinal scale. Formation of different delta lobes and shoreline changes within the Bengal Basin mostly during the Quaternary time and their controls over the geographic distributions of arsenic in groundwater are illustrated in this study.

2. Bengal Basin: an overview

The Bengal Basin, located in South Asia has been the major depocenter of sedimentary flux from the Himalayas and Indo-Burman ranges drained by the Ganges–Brahmaputra–Meghna, the largest river system in the world (Fig. 1). 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 (Uddin and Lundberg, 1998). The basin includes one of the largest delta complexes (GBM delta) in the world, covering a vast portion of the basin filled with about 5 × 10⁵ km³ of sediments (Johnson, 1994). Thick sedimentary deposits of the basin fill have been uplifted significantly along the north and eastern margins of the Sylhet trough in the northeast and along the Chittagong foldbelts of eastern Bangladesh (Uddin and Lundberg, 1998). The western part of the Bengal Basin, which covers the West Bengal of India, is mostly drained by the Bhagirathi–Hooghly river, a major distributary channel of the Ganges river (Fig. 1).

The alluvial plains of the GBM delta slope from north to south on a regional scale, but are interrupted locally by ridges and tectonically developed depressions, such as, Sylhet trough and Atrai depression. The Bengal Basin comprises of lowland floodplain and delta plain, and is surrounded by the Tertiary hills of various origins (Fig. 2; Goodbred and Kuehl, 2000; Ravenscroft et al., 2005). Within the eastern Bengal Basin, the Madhupur Tract and Barind Tract are uplifted alluvial deposits of Pleistocene age interrupt the regional surface gradient of the central basin (Morgan and McIntire, 1959). Neotectonically uplifted Lalmai Hills located to the southeast of Madhupur Tract are composed of highly oxidized clay and sand of Pleistocene age. Underneath the Pleistocene tracts, there is yellowish-brown colored sandy aquifer, formed within the Pliocene–Pleistocene Dupi Tila sand (Uddin and Lundberg, 1998).

The western part of the Bengal Basin is older than the eastern side and characterized by a sedimentary wedge of
Mesozoic to Recent age, which was deposited on a Precambrian basement that outcrops in the western margin of the area (Acharyya and Basu, 1993). The basin borders the Indian Craton (Fig. 1) to the west and once formed part of Gondwana sediments, which are still preserved in faulted troughs or grabens below the Cretaceous-Tertiary cover (Stüben et al., 2003). The Rajmahal Trap, a flood basalt unit, separates the Gondwana sequence from the younger Cretaceous-Tertiary sediments. Although the initial convergence of the Indian and Eurasian Plates began in the early Eocene time (40-41 m.y. ago), uplift of the eastern Himalayas began in the Early Miocene (Uddin and Lundberg, 1998). As a result of this convergence, the Ganges delta and the proto-Brahmaputra delta sediments were juxtaposed and gradually merged. A major part of the western Bengal Basin is now covered with Ganges–Bhagirathi–Hooghly deltaic deposits. Pre-Quaternary sedimentary deposits (mainly of Tertiary age), which are exposed in the southwestern parts of West Bengal (Fig. 2) overlie the Precambrian rocks of the Indian Craton. The aquifers that are formed by these Pre-Quaternary sediments contain groundwater with low dissolved arsenic contents (Acharyya et al., 2000).

Fig. 1. Morphotectonic map of the Bengal Basin showing the regional geologic and tectonic features. Ganges, Brahmaputra, and Meghna are the major rivers in Bengal Basin, which formed one of the largest delta systems in the world. Transect lines (A–B, C–D, and E–F) shown on this map corresponding to three regional cross-sections representing the Quaternary stratigraphy as shown in Fig. 3.
Surface elevation of the Bengal Basin is mostly less than 25 m above mean sea level, except for the Himalayan foothills in the northwest, Pleistocene tracts, and Chittagong Hills in eastern Bangladesh where surface elevations range from 25 m to about 1000 m. In the eastern GBM delta, the surface elevation is less than 15 m, with a minimum of less than 1 m in the south. Surface elevation of the GBM delta is slightly higher in the western part of the basin. Elevation is also low (<1–5 m) in the northeast Sylhet trough. The alluvial lowlands are subdivided broadly into three geomorphic regions – the Brahmaputra floodplain in the north, the Sylhet trough in the northeast, and Ganges delta plain in the south of Bangladesh (Umitsu, 1993). Landforms in the Brahmaputra floodplains are mainly characterized by natural levees, crevasse splays, alluvial sands, and channel fill deposits. Large marshes and peat lands...
characterize the Sylhet trough, which is flooded during the monsoon season by runoff from the adjoining hills. Clay, silt, silty sand, and sand form the alluvial deposits in this basin. The easternmost part of the GBM delta, which is known as the Tippera surface (Fig. 2; Morgan and McIntire, 1959), is slightly higher than adjacent floodplains, might have been tilted due to the neotectonic activities associated with Lalmai Hills, and ongoing tectonic uplift of the Indo-Burman ranges. The remainder of the delta plain is characterized by numerous river channels, abandoned channels, natural levees, broad floodplains, and marsh lands. Numerous tidal creeks and mangrove forests (Sundarbans) characterize the southern delta plain.

A simplified Quaternary stratigraphy of the Bengal Basin is summarized in Table 1. Late Pleistocene–Holocene stratigraphic units in the Bengal Basin are shown in three regional cross-sections (Fig. 3). Classification of the Late Pleistocene–Holocene units in Bangladesh (Umitsu, 1993) is correlated with the similar sequences in the West Bengal (Acharyya and Basu, 1993; Acharyya et al., 2000). The Holocene stratigraphic units are shown in two north to south regional cross-sections in Bangladesh and West Bengal in Fig. 3a–b. The lowest unit (I in Fig. 3) in the eastern Bengal Basin is characterized by a 10 m thick sandy gravel bed, whereas the similar unit (I) in the western basin consists of coarse sand (Fig. 3). Sediments in the middle unit (II in Fig. 3) are fine sand with silt and clay in the lower part, and mostly sand in the upper part and are believed to be deposited during the Early to Middle Holocene time. The uppermost stratigraphic unit (III in Fig. 3) consists of sand, silt, and clay with occasional peat layers, and was deposited during the high stand of sea level (Umitsu, 1993; Goodbred and Kuehl, 2000). A west to east cross-section (c in Fig. 3) in the coastal region of Bangladesh shows migration of deltaic lobes during the Quaternary time (Aggarwal et al., 2000). Lateral change in sedimentary facies across the basin is significant.

3. Hydrogeological and aquifer distributions

3.1. Hydrogeological conditions: eastern Bengal Basin

The deposits of thick unconsolidated Pleistocene and Holocene sediments in the Bengal Basin form one of the best groundwater productive sedimentary aquifers in the world (BGS and DPHE, 2001). Groundwater is available at shallow depths (within 10 m below surface) over most of Bangladesh from Holocene alluvium and alluvial fan deposits, floodplains and river-terraces, and the Pliocene–Pleistocene fluvo-deltaic sediments (Ahmed et al., 2004; Ravenscroft et al., 2005). Mio-Pliocene Tipam sands that form minor aquifers in the hilly areas of the northeastern and south eastern parts of the country are free of arsenic (Uddin and Abdullah, 2003). Groundwater in the Pleistocene terraces occurs below a thick clay formation, stratigraphically known as the Madhupur Clay, which varies in thickness from about 8 m to 45 m and is underlain by Dupi Tila sands of the Pliocene–Pleistocene age (Reimann, 1993). Groundwaters in Bangladesh and West Bengal in most parts are mainly dilute (total dissolved solid <1000 mg L$^{-1}$), except for the southern coastal plains and some offshore islands where the groundwaters at shallow depths are saline due to geochemical reactions with soils during recharge and leaching of salts, and mixing with seawater (Aggarwal et al., 2000). In the coastal region, fresh-water aquifers are encountered either within the first 25 m or below about 150–200 m depth (Ravenscroft et al., 2005).

Table 1
Simplified Cenozoic stratigraphy of the eastern (Bangladesh) and western (West Bengal, India) Bengal Basin (from Roy and Chattopadhyay, 1997; Monsur et al., 2001; Ahmed et al., 2004; Ravenscroft et al., 2005)

<table>
<thead>
<tr>
<th>Epoch</th>
<th>Age</th>
<th>Quaternary in Bangladesh</th>
<th>Quaternary in West Bengal</th>
<th>Lithology and sedimentology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Holocene</td>
<td>Upper</td>
<td>Alluvium</td>
<td>Alluvium</td>
<td>Clay, silt, and fine sand with occasional peat and gravels</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Matuail clay/ Basabo Formation</td>
<td>Diara Formation</td>
<td>Floodplain and deltaic deposits; mostly fine sands</td>
</tr>
<tr>
<td></td>
<td>Lower-middle</td>
<td>Chandina Formation</td>
<td>Bethuadahari Formation</td>
<td>Gray colored, fine to medium sands, with occasional coarse sands, and organic mud and peat</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dhamrai Formation</td>
<td>Sijua Formation</td>
<td>Gray colored, fine to medium sand, with clay and peat. Floodplain deposits and alluvium</td>
</tr>
<tr>
<td>Pleistocene</td>
<td>Upper</td>
<td>Kalsi bed</td>
<td>Upper Lalgarh Formation</td>
<td>Pale yellowish-brown spotted sandy clay with iron concretions, detrital nodular laterites</td>
</tr>
<tr>
<td></td>
<td>Lower-middle</td>
<td>Barind clay</td>
<td>Lower Lalgarh Formation</td>
<td>Red-brown to gray mottled clay and silt; residual deposits; Kaolinite and iron-oxides. Lalgarh contains pebble conglomerate; laterites with detrital silicified woods; highly oxidized gravels</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Madhupur clay</td>
<td>Formation</td>
<td></td>
</tr>
<tr>
<td>Pliocene</td>
<td>Upper</td>
<td>Dupi Tila Formation</td>
<td>Siwalik sediments (Pinjor/Tatrot Formation)</td>
<td>Yellowish-brown to gray, medium and coarse sand with clay; low in organic matters</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dihing Formation</td>
<td></td>
<td>Siwalik sediments consist of sand, conglomerate, and clay; deposited in fluvial environments</td>
</tr>
</tbody>
</table>
The Holocene aquifers are composed of coarse sand and gravel at the base and fine to very fine sand towards the top (Ahmed et al., 2004). Fining upward sequences are observed in the alluvial floodplain deposits throughout the basin demonstrating the shifting of the meander channels. Silt and clay predominate in the upper few meters, forming a surficial aquitard, generally less than 10 m thick (BGS and DPHE, 2001). In deltaic marshy floodplains and in central Sylhet trough, the upper aquitard is composed of thick peat and clay with silt rich in organic matter. The upper aquitard is not present in the northwestern parts of the country. Aquifers that occur along river valleys are composed of silt, fine to medium grained sand with occasional coarse sand and gravels. The Holocene aquifers in the Bengal Basin are composed mostly of gray to light gray colored sands and silts with high mica contents, often...
containing high organic matter (usually 0.5–2.5% total organic carbon in sediments) (McArthur et al., 2004). The aquifers formed by the Pliocene–Pleistocene Dupi Tila sands are tens of meters to more than hundreds of meters thick and serve as one of the best groundwater sources in the country (BGS and DPHE, 2001). Dupi Tila sand aquifers are reported only in the northern, northeastern, and southeastern Bengal Basin at approximately 50–100 m below the surface. Dupi Tila sand aquifers are not reported in the southern and western GBM delta, coastal plains and estuaries since the thickness of the overlying Holocene sediments are thicker towards the south of the basin or perhaps the Dupi Tila sediments in these areas are not oxidized and therefore are more difficult to recognize. Perhaps the Dupi Tila sediments in these areas are not estuaries since the thickness of the overlying Holocene sediments of the Ganges do not have an extensive clay layer where aquifers occur mostly under water table conditions within 150 m below surface (CGWB, 2006). Unlike the southern delta plain, many areas located to the south of the Ganges do not have an extensive clay layer where aquifers occur mostly under water table conditions within 150 m below surface (CGWB, 2006). Aquifers located between 12 m and 15 m depth below the surface in the upper delta plain are unconfined except near its southern margin, where it occurs under semi-confined to confined conditions (Das et al., 1996). Generally, there is a south-easterly gradient of the potentiometric surface sub-parallel to the general topographic slope of the area.

3.2. Hydrogeological conditions: western Bengal Basin

Hydrogeological conditions in the western Bengal Basin are slightly different from the eastern part due to regional tectonic and geologic history of the basin and precipitation pattern (i.e., Ganges catchment is much drier than the Brahmaputra and Meghna catchments). Quaternary geology of the west Bengal Basin is characterized by the shelf zone to the west of the Bhagirathi–Hooghly, the Himalayan foredeep zone to the north of Ganges and the mid-basinal zone to the east of the Bhagirathi–Hooghly (Banglapedia, 2003). Lalgarh Formation (Fig. 2) is the oldest Quaternary deposit found in southern parts of the West Bengal, which occurs at altitudes between 140 m and 80 m and comprises a boulder-conglomerate to pebble-conglomerate at the bottom, but relatively finer sediments with occasional gravels at the top (Monsur, 1995). The unit that unconformably overlies the Lalgarh Formation represents the highest and oldest alluvial terrace, and mostly comprises floodplain deposits, valley fill and delta fan complexes. Above this alluvial terrace deposit, there are sediments deposited by meandering rivers, lithologically characterized by very fine sands with silts. This unit is believed to be deposited during an Early to Middle Holocene transgressive sea level change in the Bengal Basin (Roy and Chattopadhyay, 1997). General characteristics of the Quaternary sediments of the western Bengal Basin (mostly within West Bengal) are more or less similar to those of eastern basin in Bangladesh, but the occurrences of groundwater aquifers are slightly different. The modern deltaic plain in the western Bengal Basin can be divided into two regions: the upper delta plain of meander belts of the Ganges–Bhagirathi rivers in the north; and the lower delta plain with numerous tidal creeks in the south (Das et al., 1996). The lower deltaic plain, formed in Pleistocene–Holocene time, is characterized by the presence of an extensive clay layer of varying thickness (15–76 m) which is underlain by silt, sand, and gravel (Deshmukh and Goswami, 1973). Shallow aquifer at most places in the West Bengal occurs at 12–15 m depth, while an intermediate aquifer occurs at 35–46 m. Generally, the deep aquifers in West Bengal occur at around 70–90 m depth (PHED, 1991). However, two major aquifers with depths ranging from 35 m to 46 m and 70 m to 150 m were identified in the districts of Malda, Murshidabad, Nadia, Bardhaman, and 24-Parganas (Fig. 4). Like the eastern Bengal Basin, groundwater in West Bengal occurs both under water table and confined conditions (CGWB, 2006). Unlike the southern delta plain, many areas located to the south of the Ganges do not have an extensive clay layer where aquifers occur mostly under water table conditions within 150 m below surface (CGWB, 2006). Aquifers located between 12 m and 15 m depth below the surface in the upper delta plain are unconfined except near its southern margin, where it occurs under semi-confined to confined conditions (Das et al., 1996). Generally, there is a south-easterly gradient of the potentiometric surface sub-parallel to the general topographic slope of the area.

3.3. Aquifer classification and distribution

The aquifer distributions in the Bengal Basin are poorly known because sedimentary deposits can change lithologically within a few meters and reliable borehole logs are sparse. The most commonly used conceptual models of the aquifer systems were proposed in number of studies (UNDP, 1982; MPO, 1987; Barker and Herbert, 1989). A generalized designation of aquifers defines two aquifer units: shallow aquifers (mostly alluvial and upper part of the Dupi Tila sands within a depth of 150 m) and deeper aquifers (occurring at depths below 150 m, which is locally separated from the shallow aquifer by a clay layer varying in thickness from a meter to few tens of meters) in Bangladesh (Ahmed et al., 2004). In some locations, there is a thick (~30 m) gravel bed separating the two aquifers. Based on their geological occurrence and stratigraphic positions the aquifers are also defined by sediment age: Pliocene–Pleistocene (deeper) aquifers, and Late Pleistocene–Holocene (shallow) aquifers (Uddin and Abdullah, 2003). The latter is further sub-categorized into three classes based on their relative ages in the stratigraphic column. The Late Pleistocene–Early Holocene aquifers are not always continuous throughout the country, rather they are merged and jointly form the deep aquifers of UNDP (1982) classification, and the third aquifer of Aggarwal et al. (2000) scheme. Middle Holocene aquifers may be considered similar to the main aquifer in UNDP (1982) model and the second aquifer of Aggarwal et al. (2000) model. The lower aquifer underlies the modern floodplains and deltaic plains in Bangladesh (BGS and DPHE, 2001). The Upper Holocene aquifers that are present in the deltaic plains and floodplains in the country can be correlated with
the upper composite aquifer of UNDP (1982) model and with the upper shallow aquifer (BGS and DPHE, 2001), which hosts groundwater as young as 50–100 years old (Aggarwal et al., 2000; Zheng et al., 2005).

4. Distribution of high groundwater arsenic

4.1. Geospatial distribution of arsenic

Groundwater arsenic concentration data were collected from a number of reliable sources (e.g., DPHE, 1999; BGS and DPHE, 2001; SOES, 2006; CGWB, 2006) for mapping arsenic distribution in the Bengal Basin. In Bangladesh, several large groundwater arsenic databases are available to public access (e.g., NRECA, 1997; DPHE, 1999; BGS and DPHE, 2001; BAMWSP, 2002). Among them, the National Hydrochemical Survey (NHS) database is one of the largest and complete public-domain databases. The first systematic hydrochemical survey in Bangladesh was conducted by BGS and DPHE in two phases between 1998 and 1999 covering most of the country excluding Chittagong Hill Tracts, some offshore islands, and Sundarbans mangrove forest. The hydrochemical data are available online at the British Geological Survey (http://www.bgs.ac.uk/arsenic/Bangladesh). A total of 3534 hydrochemical data were collected during the survey with their geographic locations, and analyzed for arsenic and other elements. This dataset is used in the present study for groundwater arsenic mapping of Bangladesh. However, the largest database on groundwater arsenic is available from the DPHE/UNICEF field kit survey where 29% of the tested 50,998 tubewells contain arsenic more...
than 50 μg L⁻¹. BAMWSP (2002) completed screening of more than one million wells in Bangladesh using field testing kits and 53% of the wells tested contain arsenic exceeding a concentration of 50 μg L⁻¹. In West Bengal, the largest database is created by SOES (2006), where 24% wells in a survey of total 140,150 wells contained arsenic more than 50 μg L⁻¹. In this survey, about 3.3% wells had arsenic concentration above 300 μg L⁻¹. The highest arsenic concentration in the same survey was found about 3700 μg L⁻¹ in 24 South Parganas district (SOES, 2006). The database is open to all public access at their webpage (http://www.soesju.org/arsenic/wb.htm).

Spatial distributions of arsenic in aquifers in the Bengal Basin are mapped using GIS technique (Fig. 4). Only the tubewells exceeding the arsenic concentration of 50 μg L⁻¹ are classified as contaminated and mapped in this study. In Bangladesh about 884 contaminated wells are used, whereas, in West Bengal about 111 blocks (areas covered under the jurisdiction of 111 police stations) were found to be arsenic contaminated. In those blocks the total number of arsenic contaminated wells is about 33,636. Arsenic is mostly concentrated in the central, southern, and southwestern parts of Bangladesh. High arsenic concentrations in shallow aquifers are also observed along the Ganges, Brahmaputra, and Meghna river floodplains (Fig. 4). Groundwater arsenic is mostly found in the delta plains, modern floodplains, marshes, and depressed lowlands in the Sylhet trough. About 56% tubewells in the Sylhet trough and Meghna floodplain deposits are highly contaminated (BGS and DPHE, 2001). The stable delta plains of the western part of the basin are also highly contaminated. High arsenic blocks in the western basin are mostly located to the east side of Hooghly–Bhagirathi river (Fig. 4). A few blocks located to the west of the Hooghly–Bhagirathi river are also found arsenic affected. Nine districts in West Bengal are severely affected by arsenic (>300 μg L⁻¹), whereas five districts are designated as arsenic-safe (<50 μg L⁻¹). Others are mildly affected by groundwater arsenic.

There is a strong correlation between groundwater arsenic concentration and depth, although that varies from place to place. Overall arsenic concentrations decrease with increasing well depth (BGS and DPHE, 2001). The greatest spatial variability occurs within a few tens of meters below the ground surface, and decreases significantly below approximately 100 m (Ravenscroft et al., 2005). The relationship is well illustrated in the two regional transects drawn across the basin (Fig. 5). The graphs show the groundwater arsenic concentrations are high (>50 μg L⁻¹) where well depths are relatively shallow (<50 m). In the central part of the basin (a in Fig. 5), where arsenic concentrations are extremely low, wells are apparently screened within the Dupi Tila aquifer, located beneath the Madhupur Clay unit. Well depths along this particular segment range from 25 m to 70 m below ground surface. Even within the high arsenic domain, subtle variations in arsenic concentration are noticeable, where a small change in depth significantly affects the arsenic concentrations (Fig. 5). Arsenic concentrations in the south-central parts of the basin (b in Fig. 5) show significant variations within a short lateral distance.

### 4.2. Relation between arsenic and surface elevation

There is a correspondence between increased groundwater arsenic and decreasing surface elevation in Bangladesh. The elevations of all NHS wells (BGS and DPHE, 2001) of Bangladesh are extrapolated from a Digital Elevation Model (DEM) of 300 m spatial resolution (WARPO, 2000) using the inverse distance to power gridding technique in ArcGIS 9.1 computer package. The coordinates of surveyed tubewells were used to extract elevation of each sampling location. An overlapping map of arsenic concentrations onto the DEM of Bangladesh is created (Fig. 6), which illustrates that high arsenic values correspond to low-elevation surfaces inland from the coastline. Two regional-scale transect profiles are drawn to show the hotspots of arsenic in the south-central part of Bangladesh in Fig. 7. High arsenic concentration is observed in the central part of both of these profiles where surface elevation is between 3 m and 5 m. Arsenic concentration varies from about 50 μg L⁻¹ to 450 μg L⁻¹ within the hotspot zones in both transects. Any subtle (1–2 m) variation in surface elevation is noticed to have a significant effect on arsenic variations as represented in these figures (Fig. 7). No detailed study on the relationship between surface elevation and groundwater arsenic has been conducted previously in arsenic-affected deltaic areas. A similar relationship has been reported in the Pannonian Basin of Hungary, where most of the Quaternary sedimentary aquifers are arsenic-contaminated (Varsanyi and Kovacs, 2006). In another study, Shamsudduha et al. (2006b) found that high-arsenic areas are characterized by low slopes, which can significantly affect the hydraulic gradient of shallow aquifers where groundwater flow is mainly controlled by elevation and slope variations in the recharge and discharge areas. BGS and DPHE, 2001 reported hydraulic gradients vary in Bangladesh from about 1.0 m/km in the northern part to 0.01 m/km in the south. In a different study, the hydraulic gradients have been reported as 2.0–0.5 m/km in the north, and 0.10–0.01 m/km in the south of Bangladesh (BAMWSP, 2002). Low hydraulic gradient can slow down groundwater movement significantly and reduce aquifer flushing. On the contrary, low topographic gradient can favor accumulation of finer sediments and organic matter that can drive significant microbial activities (Fe-oxyhydroxide reduction) and cause release of high arsenic in groundwater mainly at shallow depths. In contrast, in deeper tubewells in the coastal region of Bangladesh where surface elevation is low, groundwater arsenic concentrations are also low (<50 μg L⁻¹). The northeastern part of Bangladesh (i.e., Sylhet trough) is tectonically very active (Uddin and Lundberg, 1999) where surface elevation and slope is controlled by subsidence and sedimentation. In this region, high arsenic concentrations (50–250 μg L⁻¹)
are found in tubewells ranging in depth mostly from 50 m to 150 m (BGS and DPHE, 2001) where the surface elevation is less than 12 m.

5. Arsenic and Quaternary stratigraphy

The basal part of the Quaternary stratigraphy in the Bengal Basin is not easily distinguished from the older units. In the western Bengal Basin, two stratigraphic units capped by laterite or red-mottled soil profile are believed to be the representative basal unit of Quaternary stratigraphy (Niyogi and Mallick, 1973). The Dupi Tila sand and the Barind and Madhupur clays forming terraces in the eastern basin were deposited in the Early Pleistocene or earlier as found by magnetic polarity study (Monsur, 1995). The Pleistocene uplands in the western Bengal Basin were estimated to be older than 75,000 years BP based on dating of the Toba-ash beds found in these sediments (Acharyya and Basu, 1993). However, Whitney et al. (1999) estimated that geomorphic surfaces of the Barind and Madhupur Tracts dated from about 25,000 years BP to more than 110,000 years BP respectively based on $^{10}$Be isotopic analysis.

Incised alluvial valleys and lateritic uplands developed in the Bengal Basin during the last sea level low stand. During the low stand, channel incision divided the Bengal Basin into several north–south elongated highlands parallel to the main rivers – the Ganges and Brahmaputra that were flowing into the Bay of Bengal following the most direct courses while eroding the underlying Pleistocene sediments. It was not until approximately 15,000 years BP that significant sediment input was recorded on the upper Bengal Fan, apparently indicating climatic warming and increased
precipitation in the Himalayas (Goodbred and Kuehl, 2000). A gravel-rich clay layer with medium-grained sand in the western boundary of the Bengal Basin, and a similar sand layer with silt and clay from southern West Bengal have yielded $^{14}$C dates of 22,360 and 14,460 years BP, respectively (Hait et al., 1996). These radiocarbon ages correlate with the basal sand and gravel unit recorded at the Brahmaputra valley and in the Ganges delta of the eastern Bengal Basin that were believed to be deposited as entrenched valley fills during the Late Pleistocene to earliest Holocene under a low-stand setting (Acharyya et al., 2000). During that period groundwater was mainly driven by large lateral hydraulic gradient in a calm monsoonal climate, which favored extensive oxidative weathering and flushing of aquifers (i.e., Pliocene–Pleistocene aquifers) leading to the degradation of organic matter, and thus immobilizing arsenic by adsorption on amorphous Fe-oxy-hydroxides (Ravenscroft et al., 2005). Sea level rose from 18,000 to 7000 years BP and a major change in sedimentation occurred when sea level intercepted the shallow coastal platform during the Early Holocene time (Umitsu, 1993). By 10,000–11,000 years BP, a transitional phase occurred in the southern Bengal Basin when fine grained delta muds were widely deposited over the low stand oxidized and alluvial sand units (Goodbred and Kuehl, 2000). In West Bengal and Bangladesh sand sequences were deposited in the

Fig. 6. Arsenic distribution on a digital elevation model of Bangladesh shows that most of the high arsenic wells are located in low-elevated areas. High arsenic concentrations are found within the GBM delta complex and in Sylhet trough, where elevation ranges from <1 m to 10 m above mean sea level. The contour line represents arsenic concentrations of 100 µg L$^{-1}$. (Data sources: WARPO, 2000; BGS and DPHE, 2001).
central alluvial valleys followed by deposition of fine-grained sediments as the Brahmaputra flowed directly to the newly formed coastline in the south (Rennell, 1779). Sedimentation in the Sylhet trough was distinct from the greater Bengal Basin during that time. Despite relative basin deepening due to tectonic subsidence the Sylhet trough remained cutoff from the Bay of Bengal, perhaps because of a narrow Meghna river corridor that remained above sea level or had been filled with fluvial sediments (Goodbred and Kuehl, 2000). This contrast in sedimentation pattern could account for high arsenic concentrations (50–250 \( \mu g \) L\(^{-1}\)) in Sylhet region even at depths more than 100 m.

The period beginning 10,000 years BP marked an important change in the sedimentation pattern of the Ganges–Brahmaputra–Meghna delta with a rapid rise in sea level that initiated the extensive accumulation of deltaic sediments in the Bengal Basin (Goodbred and Kuehl, 2000). Sediments in the Middle Holocene unit are silty with clay in the lower part, and sands in upper parts in the eastern Bengal Basin are believed to be deposited during the Early to Middle Holocene time. The most arseniferous aquifers in West Bengal generally occur within the middle stratigraphic unit, which are composed of sand and clay and occurred at shallow depths in Malda and Murshidabad districts (Fig. 4; Acharyya et al., 2000). The uppermost

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**Fig. 7.** Relationship between groundwater arsenic and surface elevation is shown in two regional-scales transects along north–south (A–B) and west–east (C–D). High arsenic concentrations are located in topographically low areas. The profiles also illustrate that any subtle variations in surface elevation has significant effects on arsenic concentrations. Location of some rivers across the transect lines is marked.
Holocene stratigraphic unit, consisting of sandy clay, silt, and clay with occasional peat layers both in Bangladesh and West Bengal, is believed to be deposited at the beginning of 5000 years BP. During that high stand, numerous paludal basins developed within the GBM delta plains and in Sylhet trough (Umitu, 1993; Goodbred and Kuehl, 2000). Delta growth after 5000 years BP largely occurred in the eastern Bengal Basin near the modern estuary, prograding seaward through the present time (Goodbred and Kuehl, 2000). A comparative analysis of arsenic occurrences and Quaternary stratigraphic units in the Bengal Basin is presented in Table 2. Sediments of the Middle Holocene units, which were deposited dominantly under deltaic conditions, have the greatest arsenic contamination.

6. Discussion

6.1. Quaternary climate and arsenic occurrence

Late Pleistocene and Holocene sedimentation in the delta plain of the Bengal Basin, and many other alluvial basins around the world have been strongly influenced by the global changes in sea level (Bhatacharya and Banerjee, 1979; Umitu, 1993; Islam and Tooley, 1999; Goodbred and Kuehl, 2000). A positive correlation exists between location of arsenic-enriched zones within the Bengal Basin and distributions of organic-rich sediments, occurrences of marsh and peats, and Holocene sea level fluctuations (Acharyya et al., 2000; Ahmed et al., 2004; Ravenscroft et al., 2005). The Holocene transgression had direct influence over the arsenic-bearing sediments and their spatial distribution in alluvial environments in Taiwan (Liu et al., 2006). Occurrence of groundwater arsenic and its distributions within the alluvial aquifers in Red River Delta of Vietnam (Berg et al., 2001) is also believed to be associated with Quaternary sedimentation patterns linked with Late Pleistocene to Holocene sea level fluctuations.

The basal sand and gravel beds deposited in the Bengal Basin toward the end of the Pleistocene in a relatively colder and drier climatic conditions than the present-time in a low sea level stand (Banerjee and Sen, 1987). Dry climatic conditions with reduced vegetation triggered widespread and extended weathering and erosion in the Himalayas, Shillong Plateau, Rajmahal Hills, and Indo-Burman ranges causing oxidation and recrystallization of iron oxyhydroxides (Ravenscroft et al., 2005) in the upper catchment area. Over several thousands of years, relatively steep hydraulic gradients and deeper water table in the Bengal Basin extensively flushed the pre-existing sediments (e.g., Dupi Tila sands), which are free of arsenic contamination. During the low stands, restricted sedimentation mostly took place within the wide and deeper river valleys, while most sediment was drained into the Bengal fan areas further south of the present-day shelf in the Bay of Bengal (Umitu, 1993; Goodbred and Kuehl, 2000).

Between 15,000 and 10,000 years BP de-glaciation started in the high mountains with a temperature rise that
led to reinvigoration of monsoon climatic conditions in this region, and initiated a sea level rise (Goodbred and Kuehl, 2000). Meltwater from the mountains increased runoff and the major rivers deposited sediments mostly in the submarine delta, and alluvial aggradation was limited within the incised river valleys of the Bengal Basin (Ravenscroft et al., 2005). Temperature and precipitation increased during the Early to Middle Holocene time due to strong monsoon circulation in the Bengal Basin. Marine flooding led to onshore accumulation by prograding deltas after 11,000 years BP with the formation of mangrove swamps and peat basins in the delta lobes, and accumulation of organic matter in the sediment (Ravenscroft et al., 2005). The most contaminated middle units both in Bangladesh and West Bengal (Umitsu, 1993; Acharyya et al., 2000) were deposited during a high sea level condition with an initial flooding and partial sedimentation within the river valleys that led the formation of fluvial marshes, lagoons, swamps, and estuaries. High tides in the Bay of Bengal produced tidal flats and facilitated mangrove growth in brackish water that accumulated mud- and silt-rich organic material that are interbedded with lenticular sand bodies from numerous transient tributary channels (Acharyya et al., 2000). Accumulation and degradation of organic matter, formation of peat swamps, and deposition of clay materials converted the existing oxidizing environment into reducing, which supported arsenic release into groundwater mainly due to reductive dissolution of Fe-oxyhydroxides, particularly in the deltaic setting and in the swampy Sylhet trough.

Sea level rose slowly between 7000 and 5500 years BP to reach about 2–3 m higher than present-day sea level (Islam and Tooley, 1999). Southern part of the Ganges delta was invaded by the sea and numerous marine and fresh-water peat, mangrove forests, and swamps were formed in the GBM delta front. Extensive peat basins developed on the flooded coastal platform during the elevated temperature and high surface water discharge throughout the Middle Holocene time (Ravenscroft et al., 2001). Gradual migration of the Ganges river to the east and confluence with the Brahmaputra river resulted in a subaqueous delta near the modern river mouth estuary. Subdued topography resulted in sluggish groundwater flow with little flushing of the aquifer sediments of the Upper Holocene stratigraphic unit coincident with high groundwater arsenic contents (Acharyya, 2005; Ravenscroft et al., 2005).

6.2. Paleoshore events and arsenic distribution

Few authors have discussed the influence of Holocene sea level events on groundwater quality (BADC, 1992), and groundwater arsenic distribution in alluvial aquifers in the Bengal Basin (Nickson et al., 1998; Ravenscroft and Ahmed, 1998; Acharyya et al., 2000; BGS and DPHE, 2001). The Quaternary landform development in the Bengal Basin has been documented by several authors (e.g., Umitsu, 1993; Goodbred and Kuehl, 2000; Allison et al., 2003). In this study, groundwater arsenic distribution shows substantial correlations with Quaternary paleoshorelines and development of delta lobes within the Bengal Basin. A map shows the spatial distributions of high arsenic wells (exceeding Bangladesh arsenic standard of 50 µg L$^{-1}$) that are mainly located within major delta lobes restricted by paleoshorelines of the Late Quaternary period (Fig. 8). In this study, the reconstructions of paleoshorelines in the Bengal Basin are done based on physiography, lithostratigraphy, palynology, and radiocarbon age dating of sediments (e.g., Islam and Tooley, 1999; Goodbred and Kuehl, 2000). Quaternary shoreline positions during the rising sea level conditions between 7000 years BP when the maximum transgression had reached the western Bengal Basin (Umitsu, 1993) and falling sea level conditions are shown in Fig. 8. The predicted paleoshorelines at two different times (7000 years BP and 4000 years BP) clearly show that the sea level changed during the Late Quaternary time (Goodbred and Kuehl, 2000; Islam, 2001). High groundwater arsenic wells are located mostly within the central parts of the GBM delta that were significantly affected by the rising sea level conditions during the Quaternary period. Several studies suggest that the sea level rose to a maximum of about 3.0–3.5 m relative to present-day sea level during Holocene high system tracts and encroached most of the southern parts of the Bengal Basin (Islam and Tooley, 1999; Woodroffe and Horton, 2005). From digital elevation model (DEM) of the Bengal Basin, areas that are below present-day 3 m elevation are identified and the paleoshore line of Holocene high stands is drawn on the arsenic distribution map (Fig. 8). This map shows that the high arsenic concentrations (>50 µg L$^{-1}$) are located to the north of this paleoshore line. During the highest Holocene sea level conditions, the paleoshore line was probably located along this 3 m elevation line or even further north, which led to the development of extensive swamps, mangrove forestlands, and paludal basins. Ravenscroft et al. (2001) for the first time linked the development of peat land and distributions of high arsenic wells in the Bengal Basin. The formation of peat basins, related flood basins, and mangrove swamps during the Quaternary climatic change can be related to the occurrences of high arsenic in groundwaters. Aquifer sediments in these environments, which were rich in organic matter, Fe-oxyhydroxide minerals, most likely caused the occurrence of arsenic in groundwaters through reductive dissolution process governed by microbes under iron-reducing condition. Interestingly enough, groundwaters in the lower part of the GBM delta, which is presently tide-dominated, are very low (<50 µg L$^{-1}$) in arsenic concentration (SOES/DCH, 2000; BGS and DPHE, 2001). The most tube wells, located in the lower parts of the GBM delta in Bangladesh, are fairly deep (>100 m). In this coastal region, shallow tube wells are not popular because of high specific conductivities (salinity) in groundwater. About 7% tube wells, located in the tide-dominated delta bellow the latitude 22.5°N, were sampled in the National Hydrochemical Survey (BGS and DPHE, 2001) of which 60% are deeper than 100 m.
Average concentrations of As, Fe, and SO₄ in groundwaters of these coastal tubewells are 9.88 µg L⁻¹, 2.21 mg L⁻¹, and 17.49 mg L⁻¹, respectively. In contrast, the tubewells located above the latitude 22.5°N contain dissolved As, Fe, and SO₄ concentrations of about 58.32 µg L⁻¹, 3.43 mg L⁻¹, and 5.10 mg L⁻¹, respectively. The decrease in dissolved Fe, but increase in SO₄ concentrations in groundwaters of coastal region of the Bengal Basin may suggest that the low arsenic concentrations in these coastal aquifers are due to sulfate-reducing condition that limits both Fe and As concentrations by the formation of pyrite (Saunders et al., 2005). Formation of authigenic siderite (FeCO₃) in aquifers also controls the dissolved Fe concentration in groundwater (Pal et al., 2002).

Westward migration of the GBM delta is thought to influence the arsenic distributions in the Bengal Basin. Lower delta plain progradation after maximum transgression 7000 years BP can be divided into five phases with
the earliest (G1) in the westernmost delta in West Bengal formed by the Ganges (Allison et al., 2003). Different delta phases are shown in Fig. 8 with their possible ages based on clay mineralogy and radiocarbon evidence (Allison et al., 2003). The Ganges migrated eastward, occupying a series of distributaries (e.g., Gorai river) after the formation of the initial phase. The progressive infilling of the Meghna estuary in the late Holocene time is considered the last phase (GB1) of the lower delta plain formed by the Ganges and Brahmaputra rivers. The delta phase formed predominantly by the Brahmaputra and Meghna rivers (BM1) (B1 of Allison et al., 2003) has the longest development period (6000–200 years BP), and highest arsenic concentrations are found within this delta lobe in Bangladesh (Fig. 8). A deformation front (Uddin and Lundberg, 1999) is apparently located along the path of the Brahmaputra–Meghna confluence, between GB1 and G3 delta lobes. Channel shifting of the Ganges and Brahmaputra seem to have constrained within east of the deforming front. Brahmaputra was flowing east of the Madhupur terrace from about 6000–5000 years BP (Goodbred and Kuehl, 2000), depositing sediments mostly into Sylhet trough, which is also an arsenic-rich area in the country. The Ganges and Brahmaputra were flowing independently for a long period of time as shown in earlier map (Rennell, 1779), until they formed the confluence in central Bangladesh after a major avulsion completed around 1830, which initiated due to major earthquake in the year 1782 and related neotectonic activities (Goodbred and Kuehl, 2000). G1, G2, and G3 subdeltas (Fig. 8; Allison et al., 2003) that are matured and no longer prograding in the south into the Bay of Bengal are also less-contaminated by arsenic.

7. Conclusions

This study provides a comprehensive picture of groundwater arsenic in the composite Bengal Basin, combining Bangladesh and West Bengal, India. An effort has been made to describe the Quaternary geomorphic units of the complete Bengal Basin in order to link the relationship with high groundwater arsenic distributions. High arsenic concentrations are found in the tubewells located mostly in the Ganges–Brahmaputra–Meghna delta and Sylhet trough within the Middle Holocene sedimentary aquifers. Distributions of arsenic can be well linked with the Holocene sea level rise, formation of extensive peat lands, mangrove, and swampy lands in the basin. There is an inverse correlation between surface elevation and high arsenic in groundwater. Higher arsenic concentrations are found within the present-day topographically low areas that suggest the spatial distributions of higher arsenic are controlled by surface elevation and topographic slope, and groundwater gradients. During the rising and high sea level conditions around Early to Middle Holocene time, the low-lying deltaic areas in the Bengal Basin became chemically reducing that allowed arsenic to dissolve into groundwater due to the reductive dissolution of Fe-oxyhydroxide minerals under iron-reducing condition. Groundwaters in the older delta lobes in the Bengal Basin are highly affected by arsenic contamination. Delta lobes, which are in the recent past or still influenced by tidal influx, contain lower concentrations of dissolved arsenic than the relatively stable delta lobes. In addition, groundwater quality data from the National Hydrochemical Survey suggest that the coastal aquifers are in sulfate-reducing condition, which is possibly limiting the dissolved arsenic and iron concentrations in the coastal region. Compared to the severity of arsenic on millions of human lives, public domain information on arsenic in groundwater in the West Bengal is very limited and should be made abundant as in Bangladesh for future comprehensive research.

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