Title

Geological structure of an arsenic-contaminated aquifer at Sonargaon, Bangladesh

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Abstract

Continuous sediment core samples and groundwater were collected in the northern part of Sonargaon, central Bangladesh, to document the hydrogeological constraints on As-contaminated aquifers. The study area spans the alluvial plain of the Old Brahmaputra River and a Pleistocene terrace, the Madhupur Tract. The Quaternary sequence comprises Plio-Pleistocene sand, Upper Pleistocene mud, and Holocene sand units. Highly As-contaminated groundwater (50 -1000 µg/L) is found in the upper aquifer corresponding to the Holocene sand unit that underlies the alluvial plain, and plausibly appears to be closely related to the distribution of lenses of silt to fine sand. As-free (< 1 µg/L) groundwater only occurs within sediments coarser than medium sand. Highly As contaminated groundwater is characterized by low concentrations of Cl^- and SO_4^{2-} and high concentrations of NH_4^+, suggesting that the As is released in association with reduction of waters recharged during rainy season. The restricted occurrence of strongly As-contaminated (>100 µg/L) groundwater is associated with: 1) the intercalation of silt to fine sand lens in the Holocene sandy aquifer, 2) the stagnant condition of the aquifer along the buried valley, 3) the vertical infiltration of groundwater in close proximity to installed tubewells.

1. Introduction

Groundwater in Bangladesh is more strongly polluted by As than anywhere else in the world (DPHE/MML/BGS, 1999; Smith et al., 2000). Of the 6–11 million tubewells installed to depths of 10–50 m in Bangladesh, 27% exceed the Bangladesh standard for As in drinking water (50 µg/L), and 46% exceed the World Health Organization guidelines (10 µg/L) (BGS and DPHE, 2001).

The Holocene sediment is the main host of As-contaminated groundwater in the study area (e.g., Smedley and Kinniburgh, 2002; Ravenscroft et al., 2005; BADC, 1992), i.e., As-contaminated groundwater occurs in Holocene sediments along the channels of the main rivers, the Ganges, Brahmaputra, and Meghna (GBM). As-contaminated groundwater rarely occurs in the uplifted areas of Pleistocene sediment. The highest As concentration in each aquifer is commonly observed at depths of several tens of meters (BGS and DPHE, 2001).

Numerous studies have investigated the formation mechanism of As- contaminated groundwater in the Ganges–Brahmaputra–Meghna (GBM) delta, and most researchers support the mechanism that arsenic is released from iron oxyhydroxides when the hydroxides/oxides is dissolved under microbially mediated reducing conditions, as As-contaminated groundwater occurs in reducing conditions characterized by DO-free, ammonium-rich, and low-ORP chemistry, and with the presence of iron oxyhydroxide in the sediment (e.g., Bhattacharya et al., 1997; Nickson et al., 1998, 2000; McArthur et al., 2001, 2004; Tareq et al., 2003).   However, we still do not well understand the primary source(s) of arsenic and fixing process in the iron oxyhydroxide, which must be a secondary phase produced via diagenetic reaction of detrital minerals.

Detailed mapping of As-contaminated groundwater has been conducted in many areas in the country, and large varieties of As level occur in scales of a few hundred meters. Based on 6000 groundwater analyses, Van Geen et al. (2003) found that free or low As

groundwater (<10 µg/L) occures as islands a few hundred meters across in highly As-contaminated (>50 µg/L) areas of Araihazar.

Although geographical and geochemical features of the As-contaminated groundwater are well understood, information on the structure and lithofacies of the As-contaminated aquifers is limited, except large scale hydrogeology (e.g. Ravenscroft et al., 2005) and recent studies based on vertical observations of cored sediments taken from As-contaminated aquifers (e.g. AAN, 2000; Harvey et al., 2002; Anawar et al., 2003; McArthur et al., 2004). Not only the primary source(s) of As in the sediments but also the constraints on the spatial distribution of As-contaminated groundwater are not clarified until now. To obtain those solutions, we should know the lithology of aquifer sediments. Physico-chemical condition to form the As contaminated groundwater must also be related to the structure of the aqifer, since the aquifer structure affects on the pattern of groundwater flow. In the present study, we investigated the stratigraphy and structure of the aquifers at Sonargaon, located about 20 km east of Dhaka, Bangladesh, on the boundary between Pleistocene deposits and overlying Holocene alluvial deposits of the Old Brahmaputra River. The study area provides an opportunity to investigate two contrasting aquifers at depths of <60 m.

The locations of drilling sites were determined based on groundwater chemistry. Core drilling to collect sediments for chemical and mineralogical analyses was carried out at one site, and percussion drilling was undertaken at a further 11 sites. We first document the structure of the sedimentary formations, and the chemical composition of unconfined groundwater, and then show how the structure and sediment composition control the distribution of As-contaminated groundwater in this area.

2. Geomorphlogy and geology of the study area

The study area is located in the eastern part of the Bengal Basin (Fig. 1; Morgan and

McIntire, 1959; Goodbred and Kuehl, 2000; Alam et al., 2003). The Bengal Basin is the most active fluvio-deltaic system in the world, and is tectonically divided into several uplifted and subsiding areas, including the Madhupur and Barind Tracts and the Sylhet Basin (Goodbred et al., 2003). The Old Brahmaputra River runs along the eastern margin of the Madhupur Tract before joining the Meghna River in the southeast (Fig. 2). The uplifted Madhupur Tract is extensively dissected by dendritic valleys, and was uplifted during the Pleistocene along a number of NE–SW faults (Goodbred and Kuehl, 2000; Khan and Hoque, 2002). The Lower Pleistocene Madhupur Clay, which consists of silts and clays, is widely distributed throughout the upper part of the Madhupur Tract, and overlies the Plio-Pleistocene Dupi Tila Formation (Alam et al., 1990; Khan, 1991; DPHE/MML/BGS, 1999).

The study area is located on the southeastern margin of the Madhupur Tract, where reddish-brown layers of oxidized mud and sand are exposed. The eastern half of the study area is in the Old Brahmaputra floodplain. The boundary between the Madhupur Tract and floodplain trends NE–SW. The natural levees, composed of fine to medium sand with small ripples, appear as ridges along the present river and abandoned channels in the floodplain. Many settlements are located upon these levees. Under the influence of the Asian Monsoon, the floodplain is covered by water in the rainy season and cultivated in the dry season.

The Old Brahmaputra River runs N–S, with a meander amplitude of about 1 km in the center of the study area. The uplifted areas appear to be associated with neotectonism, which acted to change the course of many tributary channels during the Pleistocene (Morgan, 1970). The Old Brahmaputra River was the main flow of the Brahmaputra River until the late eighteenth century, when the main channel of the Brahmaputra River shifted to the current Jamuna channel, associated with uplift of the Madhupur Tract (Khan, 1991).

3. Methods

3.1 Core sampling and analytical methods

The subsurface geology was investigated by core drilling in December 2004 at Fatehpur Darikandi (DRK-W1, Fig. 3). The drill site was located on the edge of one of the levees. The cores (3.5 cm in diameter) were collected every 60 cm lengths continuously from the surface to 40 m depth with a split barrel sampler (Raymond sampler), and discontinuously from 40 to 80 m depth in 30–60 cm lengths at 2 m intervals. Disturbance of the cored sediment during sampling was limited, enabling the observation of sedimentary structures. To estimate the in-situ strength and density of strata, a standard penetration test (SPT; British Standard BS 1377-9:1990 and ASTM D1586) was carried out at the time of sampling. N-value measured by SPT is an indicator of the density of the ground. Soon after sampling, each lithofacies in terms of grain size, texture, structure, and fossils were visually observed in the field. The color of the samples was measured using the L*a*b* color code system with a soil color reader (SPAD-503, Minolta Co., Ltd., Tokyo, Japan). L*a*b* is developed for description of all the colors visible to the human eye by the International Commission on Illumination. The three basic coordinates represent the lightness of the color (L*, L* = 0: black, L* = 100: white), and its position between red and green (a*, negative values: green, positive values: red) and between yellow and blue (b*, negative values: blue, positive values: yellow)

The sample was then vacuum-packed together with a deoxidizer for later chemical analysis. Three plant fragments from the mud unit between 27 and 38 m depth were collected for radiocarbon dating; radiocarbon ages were determined based on ^{14}C content as measured using an accelerator mass spectrometer at the Institute of Accelerator Analysis Ltd., Kanagawa, Japan.

Reverse-circulation manual percussion drilling (the so-called "hand-flapper" or "sludger" method; Ali, 2003) was carried out at 11 sites to investigate the stratigraphy of the area (Fig. 3). In this method, the drill hole is deepened by repeatedly raising and lowering a

PVC pipe fitted with a metal crown bit (~4 cm diameter) at its base. Drilling fluid gravitationally flows into the space between the PVC pipe and the wall of the hole, returning to the surface through the pipe along with sediment cuttings. The subsurface lithology was observed from the sediment cuttings.

3.2 Determination of transmissivity

The transmissivity of each aquifer was measured via a pumping test of the three wells at the Fatehpur Darikandi site (DRK-W1, -W2, -W3). Strainers were set in the sand layer stratified between silty layers in those observation wells. The drawdown of groundwater level was measured in casing pipe with a piezometry water level logger (HOBO U20, Onset Computer Co., Ltd., USA) at 10-second intervals for 2 hours by pumping up groundwater at a rate of 22–25 L/min. Pumping flow rate is checked with measuring bucket every 10 min. The transmissivity was then calculated using the drawdown curve of groundwater level (Cooper and Jacob, 1946; Brown et al., 1963).

3.3 Water sampling and analytical methods

Groundwater samples were collected from 230 hand tubewells to characterize the chemistry of As-contaminated groundwater and thereby decide upon the locations of drilling sites (Fig. 3). Sampling was conducted in September 2003 (late rainy season) and February 2004 (late dry season) from the same wells. Temperature, pH, oxidation–reduction potential (ORP), and alkalinity were measured in the field immediately after sampling. Major element chemistry, stable isotopes, and total arsenic concentration were measured in the laboratory. In the present study, only the concentrations of total As and selected elements whose behavior is linked to that of As are described. Anions such as Cl^- and SO_4^{2-} were analyzed using ion-chromatography (Dionex IC1200). Dissolved Fe was measured by an inductively-coupled plasma atomic emission spectrometer (ICP-AES, Seiko Instruments, ICP7000). Concentrations of ammonium ions were determined by photometry using the indophenol blue

method. Arsenic concentration was determined by hydride generation atomic absorption spectrophotometry. The analytical error of all components demonstrated here is <5%, as determined from duplicate analyses of standard commercially available samples.

4. Results

4.1 Lithostratigraphy

The sediments sampled at DRK-W1 site can be stratigraphically divided into three units from the bottom to the surface of the well: the Lower Sand, Middle Mud, and Upper Sand units (Fig.4). The lithostratigraphy of the 10 percussion drill sites located in the flood plain is similar to that at the Darikandi drill site, except for site DRK-D1 where the middle mud unit is absent (Fig.5). The characteristics of each unit are described below.

4.1.1 Lower Sand unit

The Lower Sand unit comprises the deeper part of the sediment column below 38.4 m depth at DRK-W1 (Fig.4), and consists of cross-laminated medium to very coarse sand, in 1–5 m thick fining upward sequences. The lower part of each sequence consists of medium to coarse sand, including small pebbles and cross-laminations, whereas the uppermost part is formed of 1–3 m thick layers of gray silt and clay that contain parallel laminations (Table 1). The sand layers contain dominantly quartz, biotite, and muscovite. Feldspar is dominantly present in the Upper Sand, and is a minor component in the Lower Sand and the overlying Middle Mud.

Below 75 m depth at DRK-W1, the Lower Sand unit is composed of >6m very coarse sand, including granules and small-pebble-sized gravels (Fig.4). The uppermost part is reddish brown (L*: 47, a*: 8.8, b*: 16.7), and contains iron-stained quartz. The color changes gradually with increasing depth from reddish brown through yellow to light gray. The lower part of this unit (>75 m depth) is mainly composed of very coarse sand that is also reddish brown to light yellow. The color code of the sand layer ranges from L*: 40.4 to 52, a*: –0.8 to

8.8, and b*: 3.4 to 16.7 (Fig. 4,5). The SPT N-values, mostly >60, indicate dense to very dense sand, and the visual observations indicate well-consolidated mud.

The lower sand unit is exposed on the Madhupur Tract at Gulnagar (GLG-D1, Fig.3); where, the surface is oxidized and red to reddish brown (Fig. 5). Two thick clay layers are intercalated with the sand at 27–38 m depth and below 65 m at the GLG-D1 site. The lower sand unit is frequently encountered below 30–50 m depth beneath the alluvial plain, but the two thick clay layers cannot be traced beneath the alluvial plain.

The linear topographic boundary between the Madhupur Tract and alluvial plain coincides with a discontinuity in the thick clay layers within the Lower Sand, indicating a NE–SW fault downthrown to the SE.

Since the Middle Mud, deposited >54ka, unconformably covers this unit, the Lower Sand was deposited during Plio-Pleistocene.

4.1.2 Middle Mud unit

The mud unit unconformably overlies the Lower Sand, and varies from 3 to 20 m thick beneath the alluvial plain. The thickness increases towards the south (MCC-D4, Fig.5) and the north (MCC-D1, Fig.5) where the Lower Sand has been eroded. At DRK-D1, Middle Mud has been eroded. In most of the sediment columns, the upper part of the Middle Mud consists of well sorted mud, which is indicative of stable conditions during sedimentation.

The Middle Mud recovered from 28.5 to 38.4 m depth at DRK-W1 consisted mainly of consolidated silt to clay (SPT N values range from 16 to 25), including plant fragments and micaceous seams (Fig.4). The upper part (from 28.5 to 34 m) consists of clayey silt with partially developed parallel laminations. The lower part (from 34 to 38.4 m) is composed of alternating silt and fine sand, including vivianite flocs (ca. 1 mm in diameter).

The color is mainly pale gray (L*: ca 40, a*: −2.5 to −1.0, b*: −0.4 to 1.2); however, the uppermost and lowermost parts are oxidized, colored dark brown at the uppermost part (L*: 43, a*: 3.4, b*: 6.2) and gradually changing from dark brown to brown at the lowermost part toward the bottom of the unit (Table 1).

The plant fragment samples (29.6, 32.4, 33.7 m depths) contained no detectable ^{14}C (pMC < 0.12 %) and this corresponds to an age greater than 54 ka. The occurrence of vivianite flocs indicates deposition in a freshwater environment in which Fe oxyhydroxide was microbially reduced (Fredrickson et al., 1998) and the sediment was not oxidized following deposition. The SPT N-values (10-50) are lower than those for the Lower Sand.

Van Geen et al. (2003) reported a similar clay layer, with ages over 40 ka, at Araihazar, several kilometers north of the study area. They reported the clay layer, at between 15 and 30 m, separated the upper reducing from the lower oxidizing aquifers.   Based on the stratigraphic similarity of the sedimentary units and the ages of mud units (more than 50 ka), the Middle Mud in this study area corresponds to the clay layers at Araihazar. These muddy layers underlying the floodplains can be correlated with the Chandina Formation (BADC, 1992), because of similarity of these distribution and ^{14}C dating.

4.1.3 Upper Sand unit

The Upper Sand, the uppermost sedimentary unit in the alluvial plain, is exposed on the surface at all sites except Gulnagar (GLG-D1, located on the Madhupur Tract), and is typically about 25 m thick.   Thin layer <5 m thick comprising fine fraction of silt to clay is typically observed at the top of levees.   This sediment layer is lacked in two of the geological columns studied here (DRK-D1 and HHD-D1), and seems to be formed via reworking sediment from the surrounding floodplain and/or reclaiming by inhabitants.   This layer might correspond to the uppermost silt layer, regarded as the uppermost aquitard (Ravenscroft et al.,2005; Harvey et al., 2005), and 2 m thick clay/silt borrowed from the

excavated ponds to cover the villages and road (Harvey et al., 2005). However, the uppermost layer is not specified as single sediment unit and included in the Upper Sand unit in this study.

At DRK-W1, the unit is 28.5 m thick, and consists mainly of well sorted and cross-laminated fine to medium sand with small ripples. The sand is mainly composed of quartz, feldspars and micas (biotite and muscovite), and biotite becomes dominant in finer deposits (Table 1). The sediments include brown patches of Fe oxyhydroxides (goethite and/or hematite) throughout the sediment column, even in the grayish sand layer. Cyclic fining-upward sequences of <5 m thickness are occasionally observed, and thin clay to silt layers are intercalated with the sand in the upper part of the unit. The lower part consists dominantly of medium sand, which is mainly bluish gray (L*: 35 to 45, a*: –2.0 to –1.5, b*: 2.5 to 3.0), although the uppermost part (<4 m depth) is yellowish brown. The SPT N-value of the upper part ranges from 10 to 20, with occasional values of >40, indicating that the unit is poorly consolidated. The sediment at DRK-D1, fills an eroded channel to a depth of about 50 m, and rests on the Lower Sand. The Upper Sand was mainly deposited in Holocene, and forms the alluvial plain in the area.

4.2 Permeability

The transmissivity of the aquifers was determined at wells DRK-W1, W2, and W3. (Table 2). Permeabilities of each aquifer are calculated by dividing transmissivity by thickness of aquifer. The obtained values fall within the permeability of common sandy deposits (e.g., Davis, 1969; Freeze and Cherry, 1979). The permeability of the upper part of the unconfined aquifer (7.4E-03 cm/sec: 6.4 m/day, 2–12 m depth) is lower than that of the lower part (2.6E-02 cm/sec: 22 m/day, 18–28 m depth); this corresponds to the grain-size distribution, which is coarser in the lower part. Our obtained values are smaller than the previously reported values, 40-80 m/day for those of unconfined aquifers in the near area to

our study area (Ravenscroft et al., 2005; Harvey et al., 2005).   Because the deeper confined aquifer consists mainly of consolidated coarse-grained sediments, the permeability of this aquifer (1.6E-02 cm/sec: 14 m/day) is slightly lower than that of the lower part of the unconfined aquifer.

4.3 As concentration in groundwater

Fig. 6 shows the relationship between the As concentration of groundwater and well depth. Highly As-contaminated groundwater (>50 μg/L) is mostly found between 20 and 35 m depth, with the highest As concentration being >1000 μg/L. The waters taken from wells at >40 m depth are mostly free of As (<1 μg/L), while some of the deep well waters, especially in the southern part of the study area, contain high concentrations of As >50 μg/L.

The range of As concentrations and distribution pattern were the same for samples taken in both February and September. The spatial distribution of arsenic concentrations during the dry season (February 2004) is shown in Fig. 7. Most of the waters taken from wells at depths of >36 m, presumably just below the Middle Mud, have lower arsenic concentrations than those from <36 m depth. The arsenic concentrations of groundwaters beneath the Madhupur Tract are < 10 μg/L (World Health Organization guideline value), regardless of well depth.

In general, As concentrations of groundwater beneath the alluvial plain increase toward the east, with the most heavily contaminated groundwaters (>100 μg/L) found in the southeastern part of the study area, corresponding to increasing distance from the Madhupur Tract. However, the areas of less and highly contaminated groundwaters are unevenly distributed, appearing as islands (Fig. 7). The distribution of AS-free or less contaminated wells (<10 μg/L) within narrow, restricted areas, occasionally adjacent to highly contaminated wells (>50 μg/L) implies that the As contamination is controlled by local hydrogeological conditions, such as the vertical infiltration of groundwater from the surface and the lithology

of the aquifer along the flow path.

Fig. 8 shows the relationships between As and related dissolved components in unconfined groundwaters taken from <36 m depth beneath the alluvial plain. As concentrations are high in waters with little Cl^-, suggesting that As-release dominantly occurs during the infiltration of surface water, which is less polluted or less condensed by evaporation. The relationship between Cl^- and SO_4^{2-} may be explained in terms of mixtures of rainwater (free of Cl^- and with minor SO_4^{2-}) and water that is rich in Cl^- and SO_4^{2-}, as indicated by Line M in Fig. 8-a. Based on this assumption, the SO_4^{2-}/Cl^- ratio is used as an approximate indicator of reducing conditions, except for waters that contain very low concentrations of Cl^- (i.e., an SO_4^{2-}/Cl^- value above 0.8). Increasing trends in As and Fe with decreasing SO_4^{2-}/Cl^- value can support that the reductive dissolution of Fe oxyhydroxide/oxides, and consequent release of As occurs at the same time as sulfate reduction. Given the positive correlation between As and NH_4^+ concentrations, these components must behave in a similar manner; e.g., the degradation of organic matter leads to reducing conditions in groundwater to form NH_4^+ and the release of As from Fe oxyhydroxides (e.g., McArthur et al., 2004). However, reducing groundwater is not always associated with the dissolution of Fe and As, as variations in those concentrations become more pronounced with decreasing SO_4^{2-}/Cl^- values. We consider it likely that the source materials of Fe, As, and NH_4^+ are not distributed uniformly throughout the studied aquifer.

5 Discussion

Fig. 9 shows schematic profiles of the subsurface lithostratigraphy of the studied area along the lines shown in Fig. 3. The Middle Mud is a thick semi-continuous aquiclude, separates the two aquifers: the unconfined Upper Sand and confined Lower Sand aquifers. The top of the middle mud unit is almost horizontal in an E–W direction, but shows irregular undulations in a N–S direction. At DRK-D1, a buried valley filled with sandy sediments was

found, cutting through the Middle Mud and putting the upper and lower aquifers in direct contact.

Heavily As polluted groundwater (>100 μg/L) occurs in two areas, and one of those is around the DRK-D1. These sediment columns contain lenses of silt to very fine sand. Another heavily As polluted area is located between MCC-D1 and MCC-D2. Although the lithofacies at this area is unknown, as shown in a vertical N–S lithostratigraphic cross-section (Fig. 9-a), strongly As-contaminated groundwaters would be related to the local lithology such as fine-grained sediments.

In contrast, areas of free or low-As (<10μg/L) groundwater occurs as isolated islands, such as at Daulardi, the southernmost part of Mucharchar (MCC-D3), and Mamuddi (Fig. 7). It is notable that the sediment columns for these three areas (DLD-D1, MCC-D4, and MMD-D1) contain only medium to coarse sand.

Lenticular bodies with both high and low As concentrations have been reported at a scale of approximately 100 m (e.g., AAN, 2000), presumably representing palaeochannels and oxbow lakes (Ravenscroft et al., 2005). In meandering river systems, fine-grained lenses commonly form with the infill of abandoned channels, as fine particles deposited upon the quiet river floor after a change in the path of the main channel (Reineck and Singh, 1975). The As concentration of sediments is known to be high in fine-grained sediments and peat layers (e.g., AAN, 2000; Anawar et al., 2003; McArthur et al., 2004) that form in stagnant water conditions such as those found in oxbow lakes. The lower parts of abandoned channels in our study area are typically filled with coarser material, creating a zone of relatively high permeability. The absence of lenses of fine sediment in the sediment columns indicates that the low-As groundwater is sourced from a highly permeable zone within the aquifer. Our investigation demonstrates that the structure and lithology of the aquifer is an important factor in the formation of As-contaminated bodies within the sediment.

In the present study, As concentrations are higher in groundwater that is relatively poor in Cl^-. This suggests that recharge is not strongly affected by anthropogenic contamination by wastewater, or even evaporation along the flow path. At Munshiganj, 20km south of Dhaka, Harvey et al. (2002; 2005) noticed that there was convergent vertical flow into the aquifer and the young groundwater mixed with the old groundwater at about 30 m depth, where the wells were densely installed. Based on the geochemistry including radio carbon isotopes of dissolved carbon, they estimated that the As was released from the sediments associating with the recharging young groundwater. They estimated that the discharge into irrigation wells and rivers and recharge areas including ponds, rice field and rivers were spaced within 10's and 100's of meters of each other (Harvey et al., 2005). The occurrence of As contaminated and less contaminated groundwaters in our study area is concordant with their estimation. It is plausible in our studied area that As is released from the sediments in association with the downward flow of surface water recharged mainly during rainy season.

Strongly As-contaminated groundwaters in the GBM Delta share a number of common geochemical features: high HCO_3^- and NH_4^+, and low ORP, SO_4^{2-}, and NO_3^- (e.g., Nickson et al., 2000; McArthur et al., 2004; Swartz et al., 2005). High concentrations of NH_4^+ in As-contaminated groundwater have previously been explained as the byproduct of the microbially mediated decomposition of organic matter or the ammonification of organic nitrogen in peat (e.g., McArthur et al., 2004; Swartz et al., 2005). The release of As into alluvial aquifers in Bangladesh appears to follow the maturation of organic matter. The chemistry of our analyzed groundwaters (NH_4^+, As, and SO_4^{2-}) is generally consistent with those reported previously, and Fe oxyhydroxides are present as patches even in the lower part of the unconfined aquifer. Although soluble Fe does not correlate with total As in the unconfined groundwater, As release could be adequately explained by the Fe oxyhydroxides

dissolution hypothesis.

In addition to Fe oxyhydroxides, detrital Fe-rich minerals can be sources and carriers of As. In our studied sediments at DRK-W1, biotite was concentrated especially lower part of the unconfined aquifer, and >40% of total As in the whole sediment is fixed in the biotite throughout the unconfined aquifer sediment (Seddique et al., in review). The biotite is the most abundant and ubiquitous basic mineral in the GBM delta. The previous authors thought that the Fe-rich clastic minerals including biotite worked as just carriers of As since those were weathered to form Fe-oxyhydroxide (Pal et al., 2002; Harvey et al., 2005). It would be a valuable exercise to evaluate the release mechanism of As associated with the chemical weathering of detrital minerals, especially As-bearing biotite, due to the vertical infiltration of surface water into the aquifer mainly recharged at the peak of rainy season. If this is the case, biotite must be a primary source of As, and, release of this element via reduction-dissolution of Fe oxyhydroxides would be a secondary process to control As concentration in the groundwater.

The confined Lower Sand aquifer has a lower permeability than the Upper Sand aquifer. This aquifer is presumed to be recharged from the Madhupur Tract, becoming deeper toward the southeast beneath the Middle Mud unit of the alluvial plain. The uppermost sediment within this aquifer is strongly oxidized, and is gradually reduced with increasing depth. Unlike the unconfined aquifer sediments, these sediments contain less feldspars, which is the one of most easily decomposed minerals during chemical weathering. Under the oxidizing conditions observed in the Pleistocene deep aquifer of the study area, early diagenesis, including chemical weathering of detrital minerals and the decomposition of organic matter, has been completed, resulting in a relatively minor effect on As release, if present. This lower aquifer is not polluted by As or NH_4^+, except in the areas where the unconfined aquifer is heavily polluted such as the southern part of Harihardi and Temdi.

As stated above, the Middle Mud unit is absent at DRK-W1, promoting the downward migration of pollutants in association with the vertical leak of groundwater. If the excessive use of groundwater continues, As contamination will expand into the deep aquifer via vertical leakage from the unconfined aquifer.

6. Conclusion

We demonstrated the following features of the geological structure of aquifers associated with As-contaminated groundwater in the northern part of Sonargaon. In this area, arsenic pollution extends into the unconfined aquifer in association with infiltration of diluted water recharged in the rainy season. However, the distribution of highly As-contaminated groundwater (>50 μg/L) is limited to areas that contain lenses of silt to fine sand. Thus, horizontal groundwater flow has not caused significant migration of arsenic polluted groundwater. The vertical leakage of infiltrated water along outside of well pipes and/or abandoned wells, from which the pipes were removed, would cause As release into the surrounding groundwater. The source(s) of As would be locally distributed in lenses of fine sediments. Fe oxyhydroxide/oxide can be a candidate for one of the sources, however, other mineral phases, such as biotite can be another and strongly presumed one.

Acknowledgements

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Figure and Table Captions

Fig. 1   Map of the Bengal Basin (modified form Goodbread Jr. and Kuehl, 2000).

a: Modern paludal and flood basins, b: Modern floodplain and delta plain, c: Holocene uplift,

d: Pleistocene uplands, e: Tertiary uplands.

Fig. 2   Location map of the study area.

Fig. 3   Locations of sampling wells and drill sites.

Fig. 4   Drill log and color indices of sediments at the Darikandi drill site (DRK-W1).

SPT N-values indicate the stiffness of the strata with blow counts of a 140 lb. hammer during

penetration sampling. L*a*b* shows the color code of the strata. L*: lightness coordinate,

representing the range from black to white; a*: red/green coordinate, with +a* indicating red

and −a* indicating green; b*: yellow/blue coordinate, with +b* indicating yellow and −b*

indicating blue. The location of the drill site is shown in Fig. 3.

Fig. 5   Drill logs compiled during percussion drilling in the study area.

The locations of the drill sites are shown in Fig. 3.

Fig. 6   Arsenic content of groundwater as a function of depth.

Fig. 7   Maps of the spatial distribution of the arsenic content of groundwater.

Fig. 8   Plots of major chemical compositions, As and its related components.

(a) Cl^- - SO_4^{2-}; (b) Cl^- - Total As; (c) SO_4^{2-}/Cl^- - Total Fe; (d) SO_4^{2-}/Cl^- -Total As; (e) Total Fe - Total As; (f) NH_4^+ - Total As

Fig. 9   Geologic cross-sections across the study area.

(a): N–S section, (b): E–W section. The locations of the cross-sections are shown in Fig. 3.

Table 1   Lithology of deposits in the study area.

Table 2   Results of pumping tests and obtained coefficients of permeability for the upper and lower sand units.
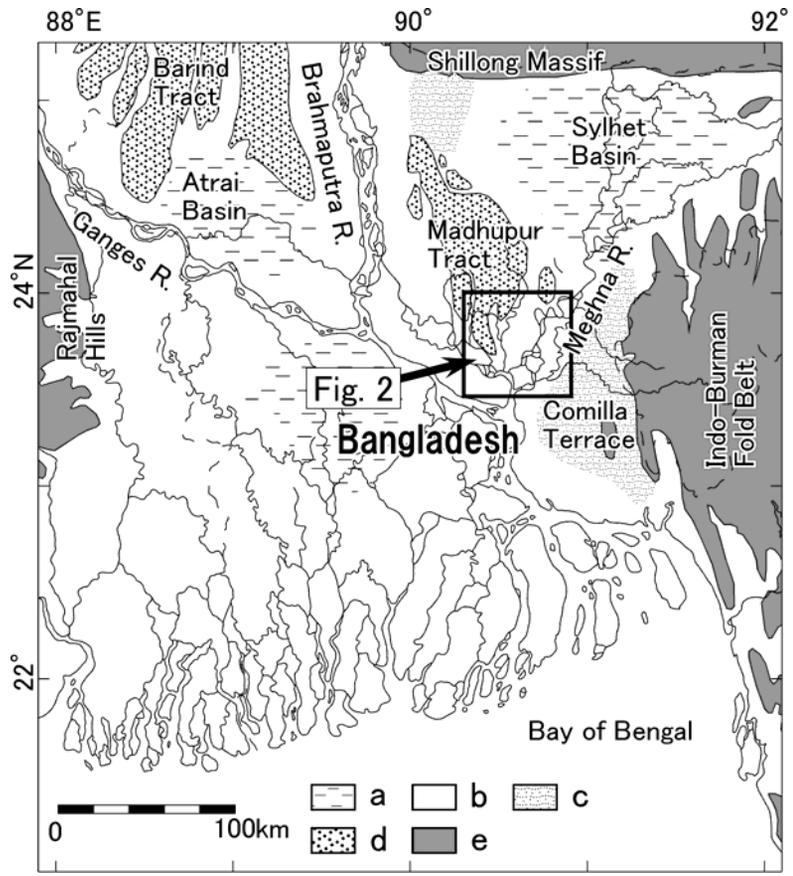
Table 1 Lithology of deposits in the study area.

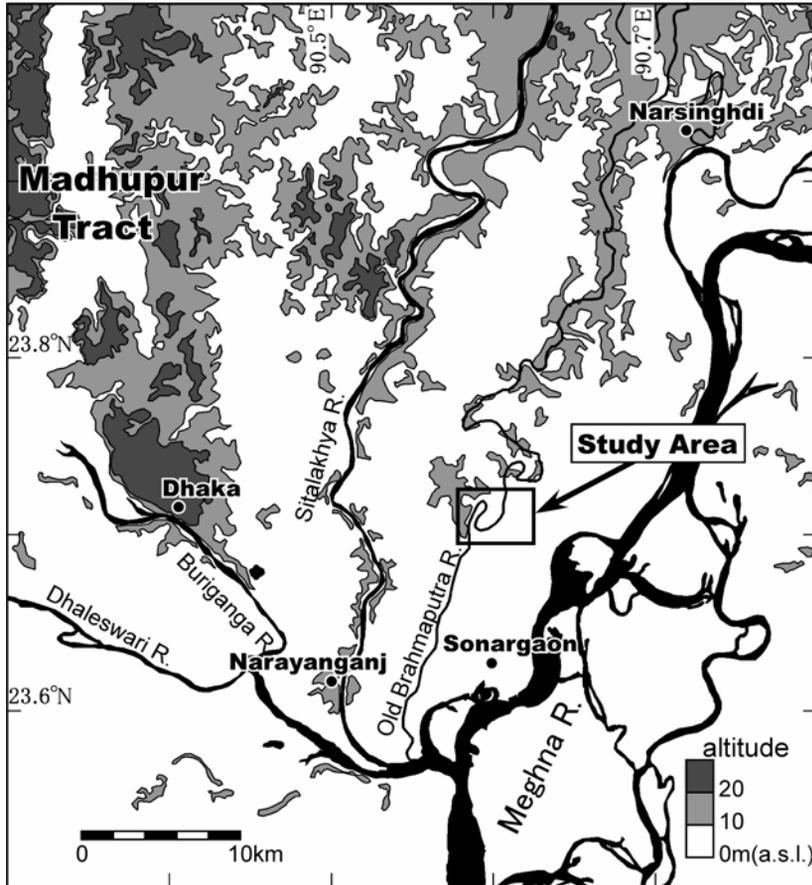| Unit	Lithology	Remarkable minerals	Color		
			L*	a*	b*
Upper Sand	fime to medium sand	quartz, feldspar, biotite, muscovite	35 to 45	-2.0 to -1.5	+2.5 to +3.0
			Munsell:5B6/1-5/1,10BG5/1 (2.5y5/2-5y5/1)		
Middle Mud	silt to clay intercalating thin fine sand	biotite, muscovite, vivianite	40 (43)	-2.5 to -0.4 (3.4)	-0.4 to +1.2 (6.2)
			Munsel: 5B/6/1-5/1,10BG5/1 (5YR2/3)		
Lower Sand	medium to coarse sand intercalating silt and clay	iron-stained-quartz, biotite, muscovite	40 to 52 (47)	-0.8 to +8.8 (+8.8)	+3.4 to +16.7 (+16.7)
			Munsel: 5B/6/1-5/1 (5YR3/3)		

(color code of upper most part)

Table 2　Results of pumping test and coefficient of permeability
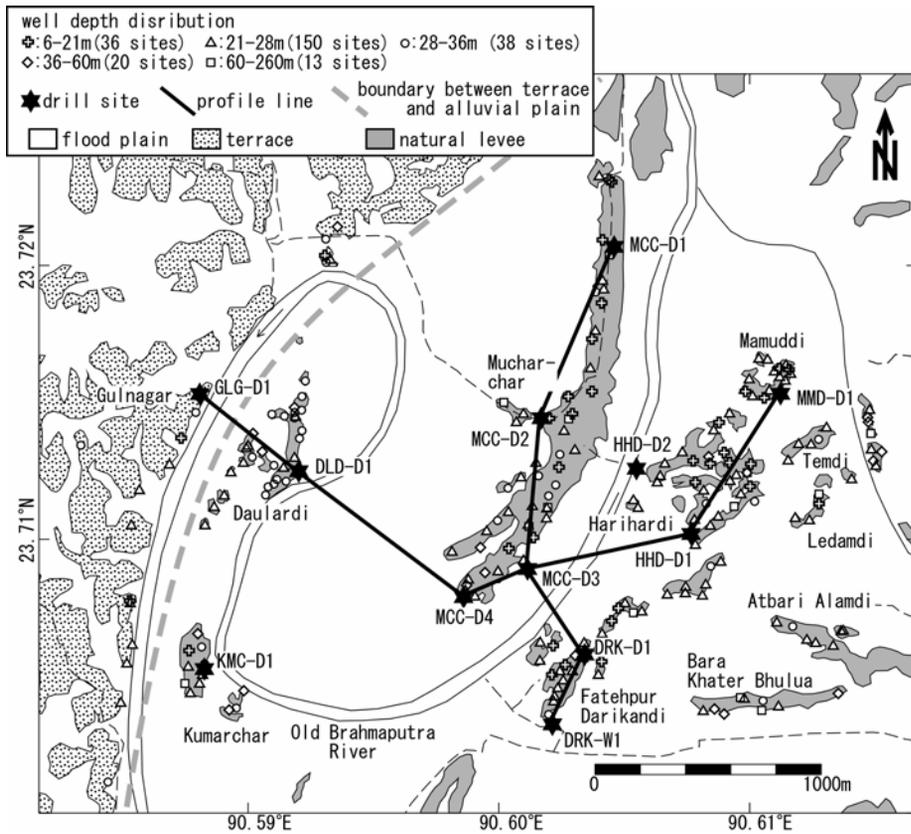in the Upper and Lower sand units

Parameter	Upper sand unit (un confined aquifer)		Lower sand unit (confined aquifer)
	upper part	lower part	
Flow rate of pump up (cm^3/sec)	367	400	413
Aquifer thickness (cm)	1000	1000	1500
Drawdown rate (cm/log$_{10}$section)	9.15	2.80	3.20
Transmissivity (cm^2/sec)	7.35	26.19	23.58
Coefficient of permeability (m/sec)	7.4E-03	2.6E-02	1.6E-02

Mitamura et al., Fig. 1

Mitamura et al., Fig. 2

well depth disribution
⊕:6–21m(36 sites)　△:21–28m(150 sites)　○:28–36m (38 sites)
◇:36–60m(20 sites)　□:60–260m(13 sites)

★drill site　　／profile line　　boundary between terrace and alluvial plain
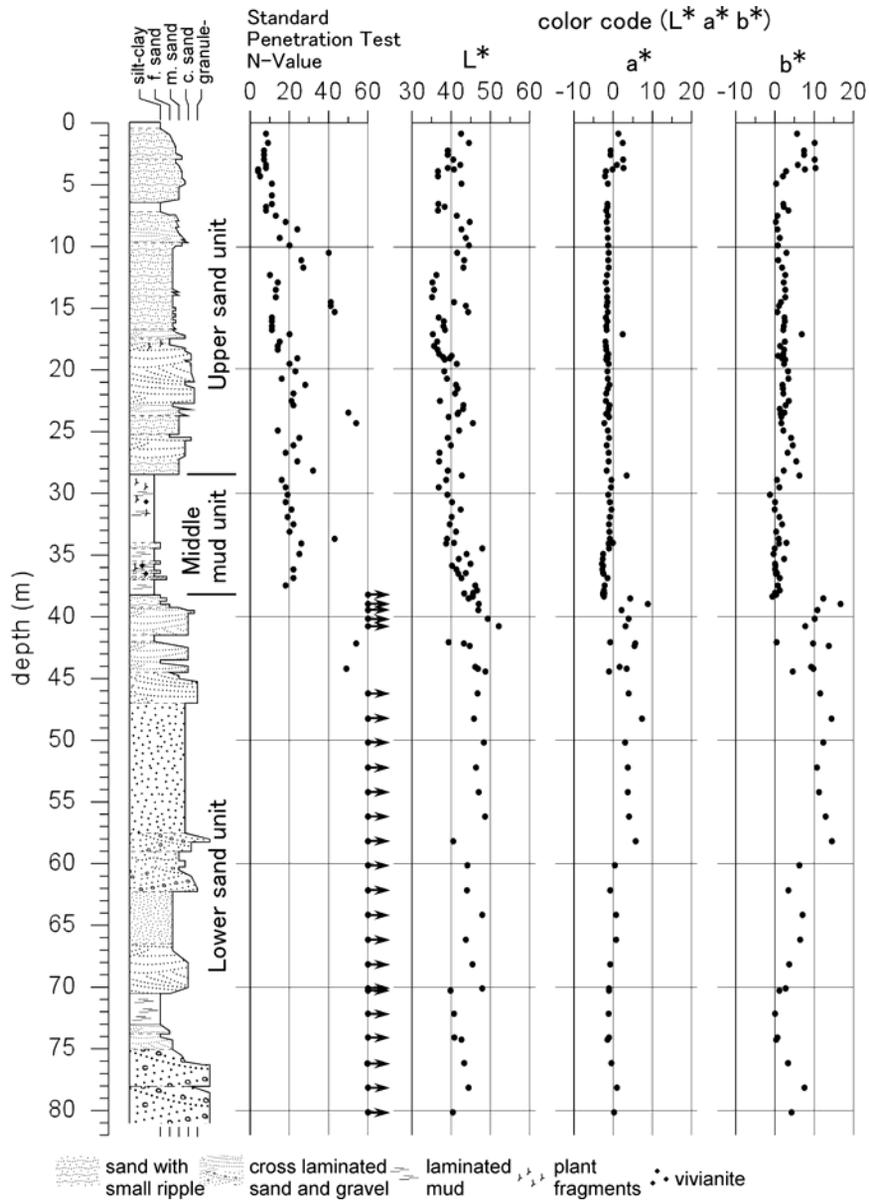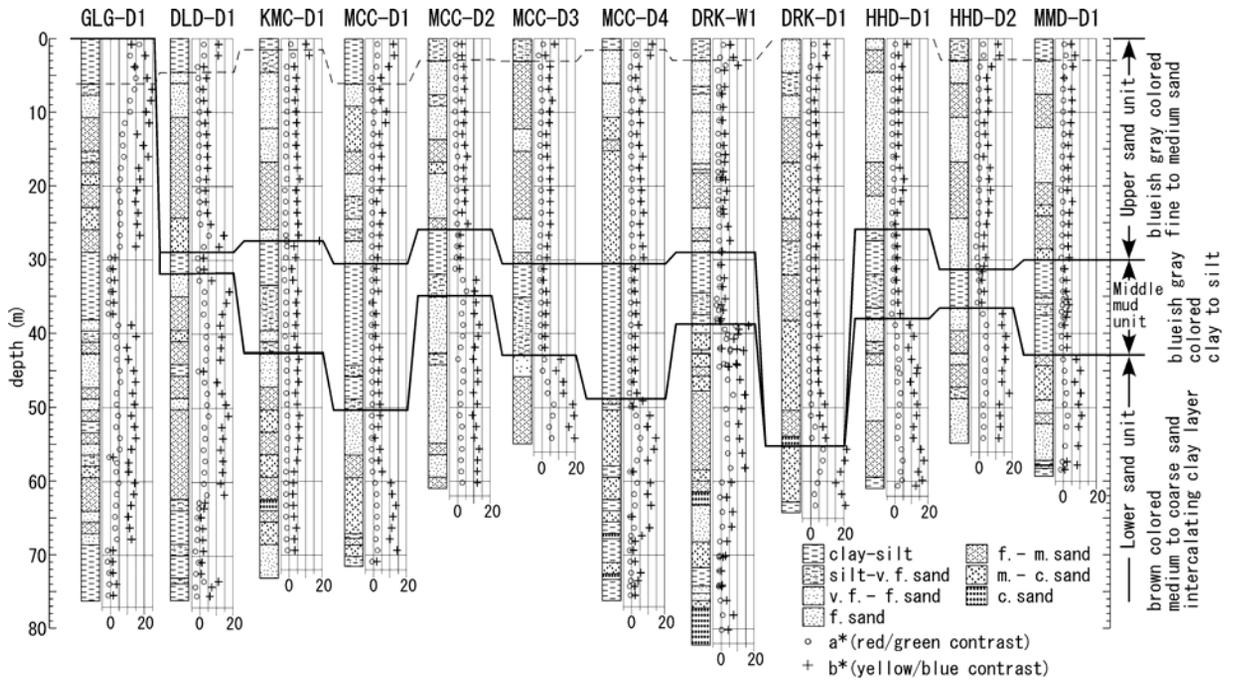
☐flood plain　▨terrace　▨natural levee
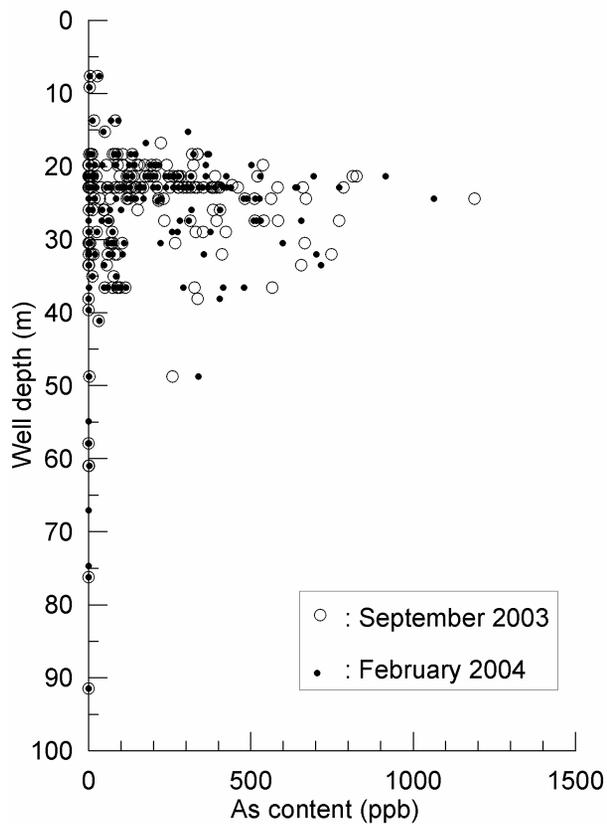
Mitamura et al., Fig. 3
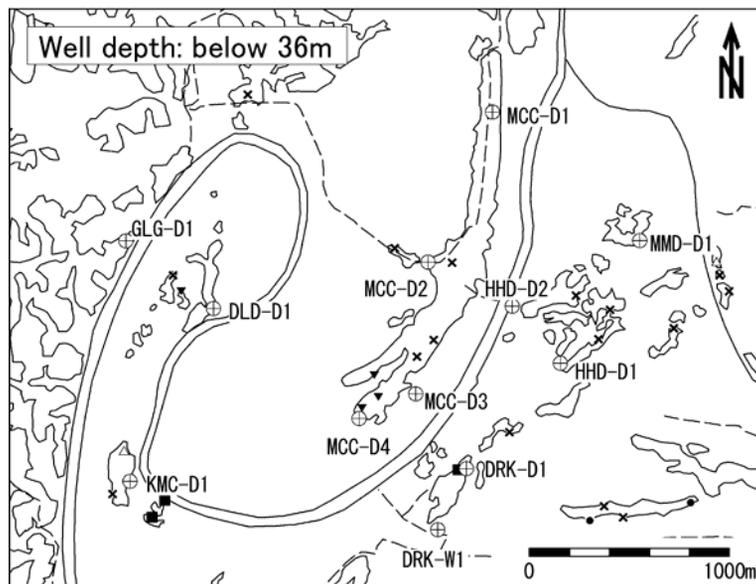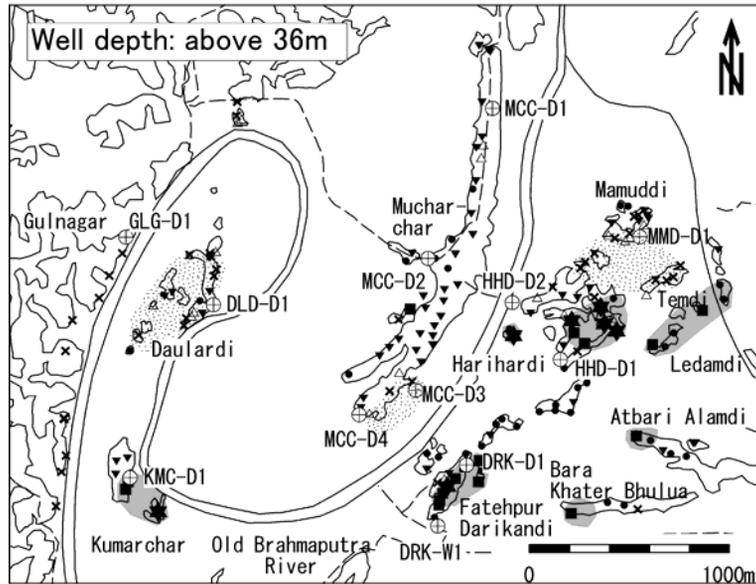
Mitamura et al., Fig. 4

Mitamura et al., Fig. 5

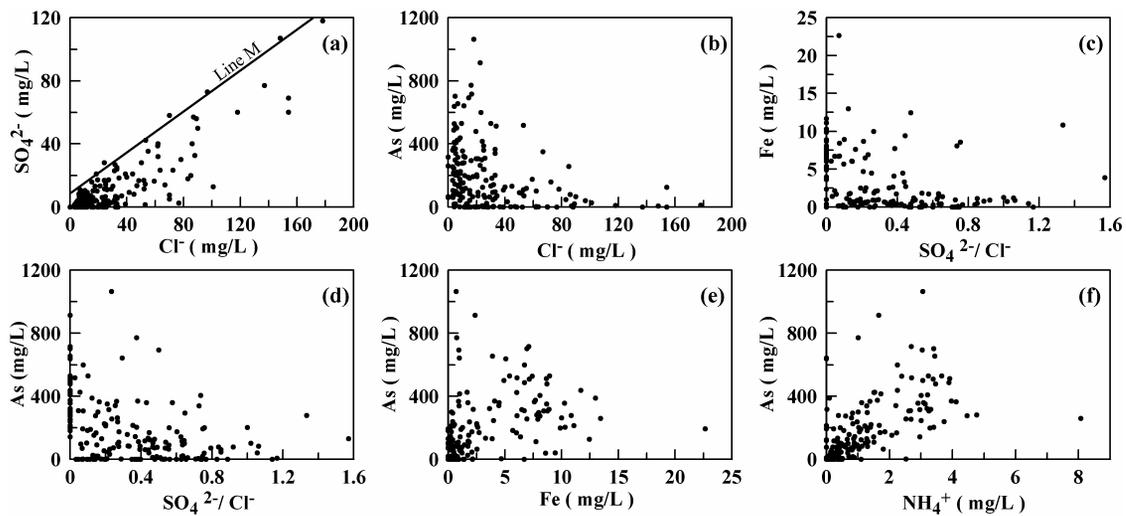Mitamura et al., Fig. 6

Well depth: above 36m

N

MCC-D1

Mamuddi

Muchar-
char

Gulnagar GLG-D1

MMD-D1

MCC-D2

HHD-D2

Temdi

DLD-D1

Harihardi

HHD-D1

Ledamdi

Daulardi

MCC-D3

Atbari Alamdi

MCC-D4

KMC-D1

DRK-D1 Bara
Khater Bhulua

Fatehpur
Darikandi

Kumarchar   Old Brahmaputra
River

DRK-W1

0   1000m

Well depth: below 36m

N

MCC-D1

GLG-D1

MMD-D1

DLD-D1

MCC-D2

HHD-D2

HHD-D1

MCC-D3

MCC-D4

KMC-D1

DRK-D1

DRK-W1

0   1000m
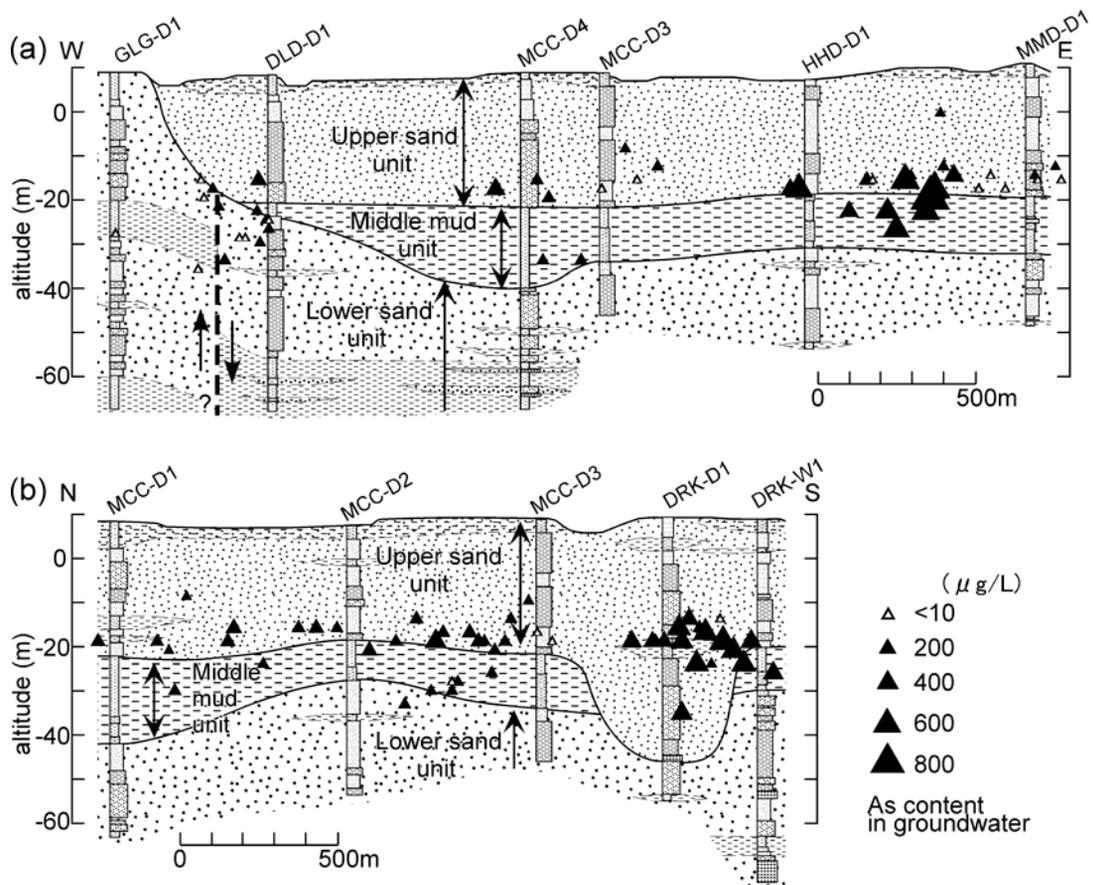
As content in groundwater (μg/L)
✕ 0–10   △ 10–50   ▼ 50–200   ● 200–400   ■ 400–600   ✦ 600–800

Mitamura et al., Fig. 7

Mitamura et al., Fig. 8

(a) W GLG-D1 DLD-D1 MCC-D4 MCC-D3 HHD-D1 MMD-D1 E

Upper sand unit

Middle mud unit

Lower sand unit

altitude (m)

0 500m

(b) N MCC-D1 MCC-D2 MCC-D3 DRK-D1 DRK-W1 S

Upper sand unit

Middle mud unit

Lower sand unit

altitude (m)

0 500m

(μg/L)
△ <10
▲ 200
▲ 400
▲ 600
▲ 800

As content in groundwater

Mitamura et al., Fig. 9