Arsenic toxicity of groundwater in parts of the Bengal basin in India and Bangladesh: the role of Quaternary stratigraphy and Holocene sea-level fluctuation

S.K. Acharyya · S. Lahiri · B.C. Raymahashay · A. Bhowmik

Abstract Arsenic toxicity in groundwater in the Ganges delta and some low-lying areas in the Bengal basin is confined to middle Holocene sediments. Dissected terraces and highlands of Pleistocene and early Holocene deposits are free of such problems. Arsenic-rich pyrite or other arsenic minerals are rare or absent in the affected sediments. Arsenic appears to occur adsorbed on iron hydroxide-coated sand grains and clay minerals and is transported in soluble form and co-precipitated with, or is scavenged by, Fe(III) and Mn(IV) in the sediments. It became preferentially entrapped in fine-grained and organic-rich sediments during mid-Holocene sea-level rises in deltaic and some low-lying areas of the Bengal basin. It was liberated subsequently under reducing conditions and mediated further by microbial action. Intensive extraction of groundwater for irrigation and application of phosphate fertilizer possibly triggered the recent release of arsenic to groundwater. This practice has induced groundwater flow, mobilizing phosphate derived from fertilizer, as well as from decayed organic matter, which has promoted the growth of sediment biota and aided the further release of arsenic. However, the environment is not sufficiently reducing to mobilize iron and arsenic in groundwater in the Ganges floodplains upstream of Rajmahal. Thus, arsenic toxicity in the groundwater of the Bengal basin is caused by its natural setting, but also appears to be triggered by recent anthropogenic activities.

Key words Arsenic in groundwater · Bangladesh · Bengal basin · Deltaic sedimentation · Holocene sea-level rise · West Bengal

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S.K. Acharyya (✉) · S. Lahiri
Geological Survey of India, 27 Jawaharlal Nehru Rd. Calcutta, 700016, India
e-mail: skacharyya@yahoo.com
Tel.: +249-6976(0), 466-0215(R)
Fax: +249-6956

B.C. Raymahashay · A. Bhowmik
Department of Civil Engineering,
Indian Institute of Technology, Kanpur 208016, India

Introduction

Arsenic toxicity in groundwater in southern West Bengal (India), and southern and eastern parts of Bangladesh, is an alarming environmental problem that was first recognized about a decade ago. The presence of arsenic in groundwater at concentrations that exceed the permissible limit of 0.05 mg/l (WHO 1993, provisional specification <0.01 mg/l) for human consumption was recorded in West Bengal in 1978, and the first case of arsenic poisoning was diagnosed in 1983. Within a span of 12 years, over 3 to 4 million people in six districts in southern West Bengal, which is located east of the Bhagirathi River (Fig. 1), were found to be affected by arsenic poisoning in groundwater. In Bangladesh, arsenic contamination of groundwater was first detected in 1993. The widespread malady with “no equal in medical history” was soon reported in many parts of southern Bangladesh, which confirms arsenic contamination in shallow and deep tube wells, as well as cases of arsenicosis. In terms of the numbers of people exposed, it is the most serious groundwater arsenic problem in the world. It is an irony that arsenic toxicity from West Bengal and Bangladesh was recognized only after extensive exploitation of groundwater for irrigation and drinking water, and after incurring huge investments. Application of groundwater irrigation for high yielding ‘Boro’ rice and a switch to multi-crop practices have been responsible for achievements in food-grain self-sufficiency, and a reduction in infant mortality from diarrheal diseases as a result of piped water supplies. The prime question is whether this arsenic toxicity problem was inherent in the groundwater

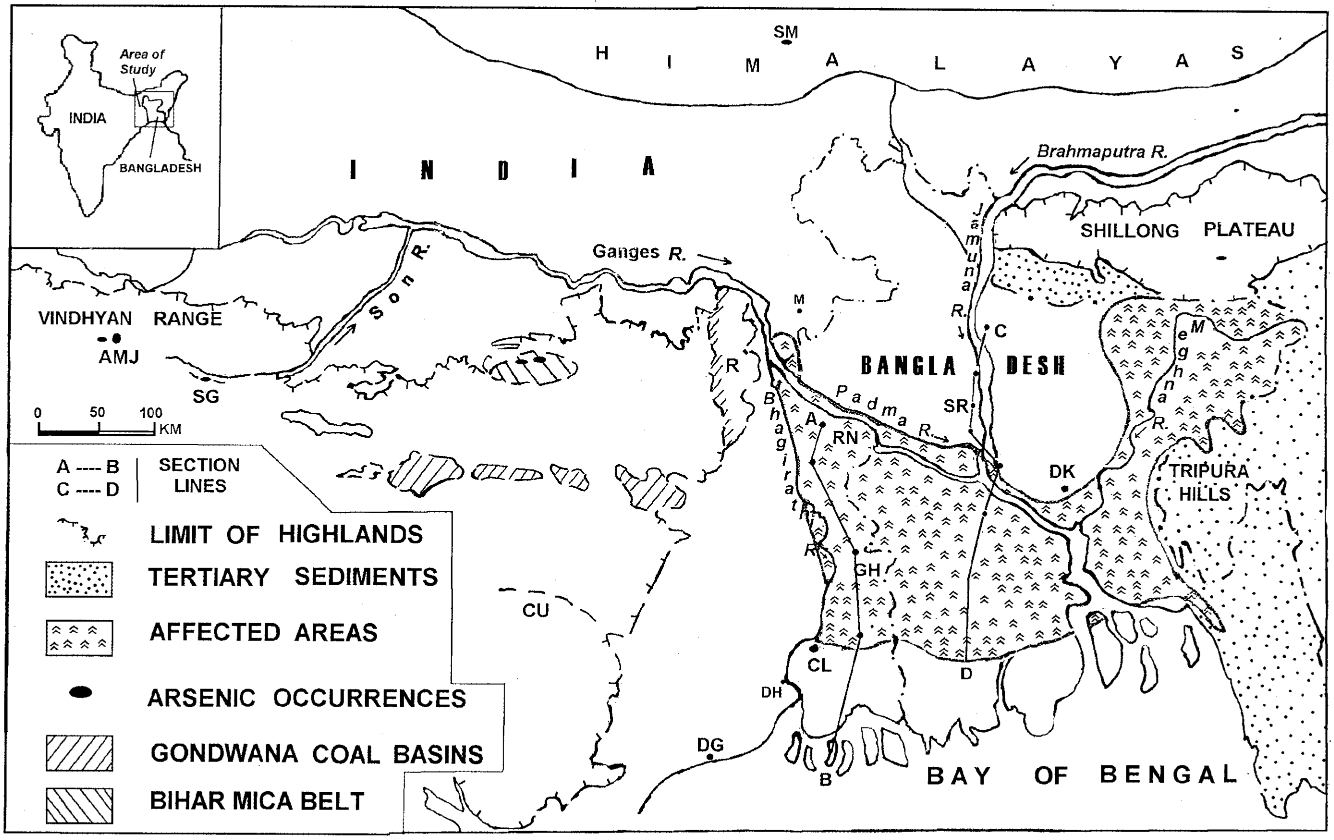

Fig. 1
Map of arsenic-affected areas in parts of Bengal basin and potential sources. *AMJ* Amjhore pyrite mine, *CL* Calcutta, *CU* Copper belt Bihar, *DH* Diamond Harbour, *DG* Digha, *DK* Dhaka, *GH* Ghetugachi, *M* Malda, *R* Rajmahal hills, *RN* Raninagar, *SG* Son valley gold belt, *SM* Samthar, *SR* Sirajganj

from the affected area or whether it was triggered by recent anthropogenic activities?

Unlike other parts of the world, the arsenic-affected area in the Bengal basin has an extensive span of the alluvial and deltaic tract, which is free of volcanic activity or thermal springs, and virtually devoid of industry and mining activity (Welch and others 1988; Wang and Huang 1994; Smedley and others 1996). The fact that the affected area is mainly located in the delta of the Ganges, Brahmaputra and Meghna rivers suggests multiple natural sources for the arsenic.

There are two opinions: (1) the cause is entirely natural or that (2) it is partly anthropogenic, and is triggered by intense exploitation of groundwater, application of fertilizer and pesticides.

Setting and distribution of the arsenic-affected area

The arsenic-affected area, as initially recognized in West Bengal and Bangladesh, is mostly confined to the lower flood-delta plains of the Ganges River, downstream of the Rajmahal Hills (Fig. 1). The Gangetic alluvial tract upstream of the Rajmahal Hills, in the states of Bihar and Uttar Pradesh (UP), in India, is generally free of arsenic toxicity. However, the adjacent part of the Ganges delta in Bangladesh is equally affected in the Bengal basin. It was soon further recognized that arsenic toxicity in Bangladesh extends also to the Sylhet basin and flood-delta plains of the River Meghna in eastern and southern Bangladesh (Fig. 1). Of these affected areas, groundwater from the Ganges delta has been more closely studied, and a large variability in spatial distribution and content of arsenic has been established at local and regional scales. However, there is a strong correlation between the arsenic-affected area and the geomorphology and Quaternary geology of the Bengal basin (Figs. 1 and 2). A brief description of the regional setting of the Ganges delta and the geomorphology of its surrounding region is thus relevant.

The Rivers Ganges and Brahmaputra flow into their combined deltaic basin from the north-west and the north, whereas the Meghna River drains into the delta from the north-east (Fig. 2). The lowland alluvial basin of the Rivers Ganges, Jamuna and an older Brahmaputra course have been entrenched in Pleistocene terraces. Hills of older rocks and uplands comprised of a laterite plain and an older alluvial plain (OAP) (Niyogi 1975, Bhatacharya and Banerjee 1979), of Pleistocene and early Holocene age, flank the western side of the lowland delta in West Bengal (Fig. 2). The term OAP is used here specifically

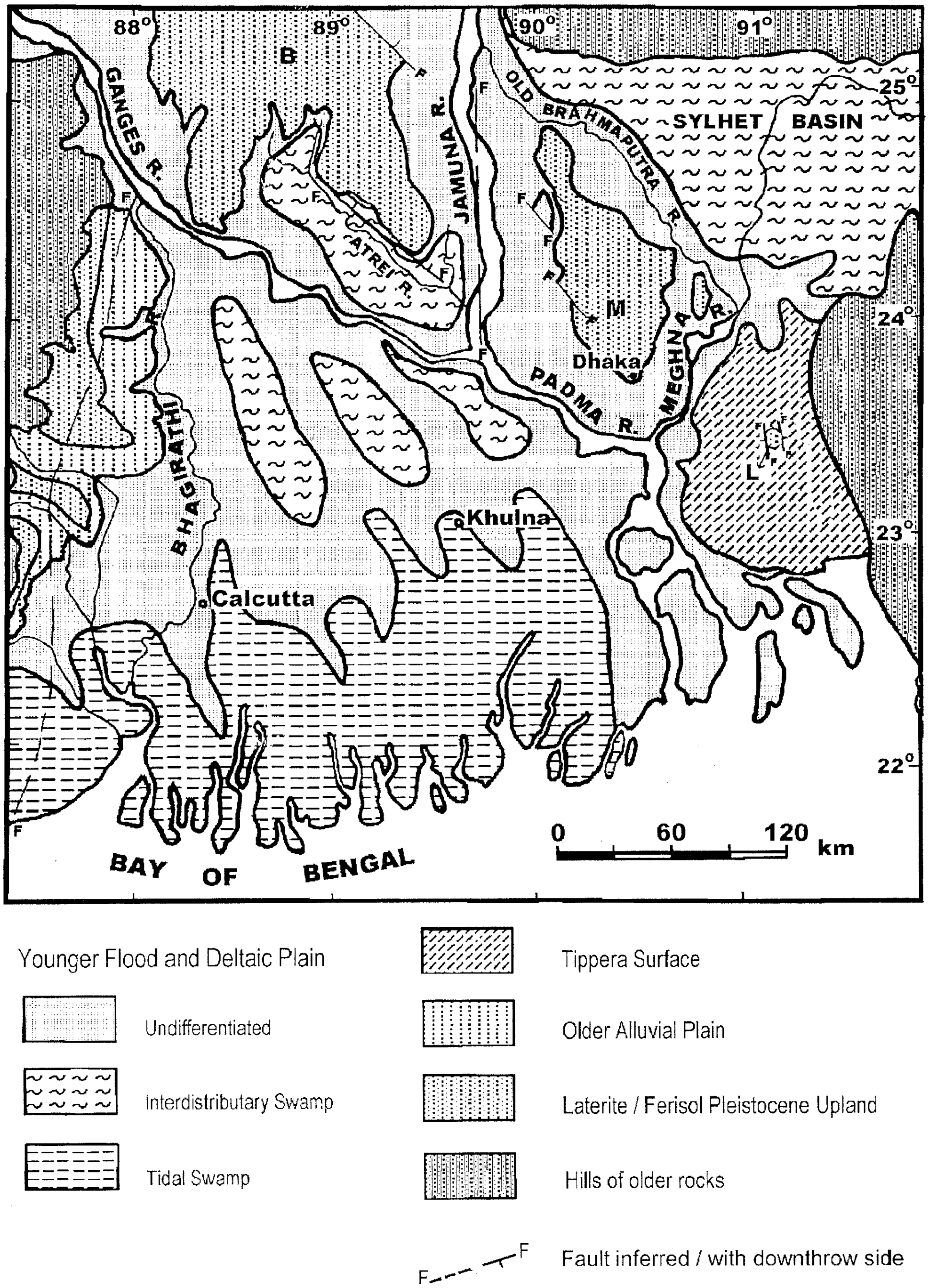

Fig. 2
Landforms and depositional environments in parts of Bengal basin. *B* Barind, *M* Madhupur, *L* Lalmai (Pleistocene uplands)

for the lower upland, although both the upland plains are older alluvial plain in nature. The River Bhagirathi, a prominent tributary of the Ganges delta system, has scoured its younger and recent deltaic basin over the eastern flank of the OAP. The Damodar River had also formed two alluvial fans (Deshmukh and others 1973), which are younger than OAP, but are truncated by the younger delta plain (YDP) of the Bhagirathi River system. The Damodar River was flowing in an easterly direction to meet the Bhagirathi during the middle of 18th century, but has since shifted its river mouth 128 km to the south. The arsenic-affected aquifers in West Bengal occur beneath the lower-level flood-delta plain of the Ganges River system. The bordering western upland terraces and the Damodar fans are free of arsenic problems (Mallick and Rajgopal 1996).

The Pleistocene terraces to the north and in the central part of the lowland delta in Bangladesh are called the Barind and Madhupur tracts (Morgan and McIntire 1959). The combined delta is flanked to the east by the Tertiary hills. The south-eastern part of the delta, which is comprised of the Tippera surface (Morgan and McIntire 1959), is slightly elevated tectonically and within it occurs the popped-up Pleistocene beds of the Lalmai hills

(Fig. 2). The elevation of south Bengal lowland, in West Bengal and in Bangladesh, is mostly <24 m above sea level (m a.s.l.) in the west and north, and most of the southern part of the region is lower than 2 m a.s.l. The relative height of the Pleistocene upland above the surface of alluvial lowland is 3–15 m in the west and north, and 0–5 m in the south. The alluvial lowland may be subdivided into the Ganges delta to the south, the Brahmaputra-Jamuna floodplain in the centre and the Sylhet flood basin in the north-east (Fig. 2). The Ganges delta is located to the south of Barind and Madhupur upland. The Brahmaputra-Jamuna floodplains are located between the Barind, Shillong Plateau and Madhupur uplands, whereas, the Sylhet flood basin is surrounded by the Shillong Plateau, the Madhupur upland and the Tertiary hills of Tripura. The Old Brahmaputra River, a former trunk river channel of the river, and the Meghna River, join to the south of the Sylhet basin. The elevation of the central Sylhet basin is lower than 3 m a.s.l. and, thus, most of the basin is marshy and covered by flood water every season. Extending towards the Bay of Bengal, the combined deltaic complex has numerous abandoned tributary channels and stream scars. The entire zone is a maze of interconnecting abandoned channels and tributaries, which do not cluster into deltaic lobes. Broad expanses of tidally deposited mud and silt overlap the seaward sloping deltaic plain (Fig. 2).

The tectonics of the Bengal basin is revealed by the presence of several normal faults that were active during the Quaternary (Fig. 2). The Madhupur upland is bordered to its west by a series of en echelon faults, and the Jamuna, Padma and Atrai River courses are also controlled by faults (Morgan and McIntire 1959; Alam and others 1990). The western margin of the Bengal basin is affected by a basin marginal fault that was active during the Cenozoic (Mukherjea and Hazra 1997), whereas its northern faulted boundary against the Shillong Plateau has been active since the late Cenozoic (Johnson and Alam 1991). It has been inferred that many tributary channels in the lower delta complex were abandoned because of sudden gradient changes that were a result of neotectonism (Morgan 1970).

In Bangladesh, the older oxidized residual clay and sediments beneath the Barind and the Madhupur uplands to the north are free of arsenic problems, whereas the lower plains of the Ganges delta are affected. The braided river and floodplain deposits of the Brahmaputra-Jamuna are unaffected. However, the Sylhet basin and Meghna floodplains are affected, and the most affected aquifer lies beneath the Ganges-Meghna delta mouth and the Tippera surface of south-east Bangladesh (Anonymous 1999). The oxidized Pleistocene sediments from the Lalmai horst block, located within the latter belt, are free of arsenic problem. Thus, it is observed that the arsenic-affected sediments in the Bengal basin are controlled by geomorphology and Quaternary stratigraphy.

Quaternary stratigraphy and lithology: the position of arsenic-contaminated aquifers

An attempt has been made to broadly identify the stratigraphic level of arsenic-toxic aquifer zones that occur beneath the alluvial plain of the south Bengal basin. The sediments from the arsenic-free western uplands, which are comprised of the laterite plain and older alluvial plain, and that flank the lower level younger flood-delta plain of the Bhagirathi-Ganges Rivers in West Bengal, are not yet well correlated and dated. In addition to their stepped morphological character, the relative antiquity of deposits from these uplands have been worked out from their state of preservation, and the nature and thickness of the developed soil profiles (Niyogi 1975; Bhatacharya and Banerjee 1979). Dendritic valley cuts that are filled with younger flood-delta plain sediment penetrate as fingers into the older alluvial plain. Likewise, valley-cut fingers with older alluvium penetrate into the lateritic plain. The nature of the palaeochannels and alluvium from the older alluvial and laterite plains, are atypical of deltas, although some researchers disagree (Niyogi 1975; Bhatacharya and Banerjee 1979). The lateritic and/or ferrisol profile in the laterite plains is 10–20 m thick. Double profiles of laterite have been recorded at many places. A laterite/ferrisol profile has developed on Quaternary alluvium that contains abundant lateritic pisolites; these are derived from the adjacent provenance area capped by an in situ laterite profile. A calcareous concretionary clay soil is 2–5 m thick in OAP, and contains a top section of 0.1–1 m of dark brown soil with soft Fe-Mn concretions and an absence of calcareous concretions. Soil on the young delta plain (YDP) is 1–2 m thick with a top section of up to 0.2 m, with soft Fe-Mn concretions and calcretes that are locally present in the lower section. Soil formation is practically absent in the recent delta plain.

Based on integrated analysis of several borehole records from West Bengal, which is part of the Bengal basin, the subsurface continuity of these morphostratigraphic units have been established (Niyogi 1975; Bhatacharya and Banerjee 1979), whereas, lower subsurface units from YDP may not have outcropped. The presence of characteristic soil profiles, e.g. laterite/ferrisol and calcrete, which is recognized at subsurface, has helped such correlation. The base of the Quaternary section was difficult to recognize, but in many boreholes a sequence of mostly clay and sand that have formed in saline water and locally contain microfossils has been assigned to an Upper Pliocene age (Sengupta 1966). The laterite plain morphostratigraphic unit has been linked to one or two subsurface stratigraphic units that are capped by laterite or have a red-mottled soil profile that overlies the eroded Pliocene beds (Niyogi and Mallick 1973; Bhatacharya and Banerjee 1979). Beneath their laterite/ferrisol profiles, these units are represented by coarse alluvium and often have upwards fining sequences. An unoxidized calcrete-bearing and relatively finer alluvium unit overlies these, which has been correlated to the OAP morphostratigraghic unit (Niyogi 1975; Bhatacharya and Banerjee 1979). The Quaternary sequence developed along the western flank of the Bengal basin usually has a wedge-like development and thickens eastward, and usually has a greater number of units.

It is pertinent to point out that the presence of the youngest Toba-ash bed marker (75,000 years b.p.) has been recorded in the basal parts of the Quaternary profile from the Brahmani and Barakar River sections, slightly to the west of the Bengal basin margin (Acharyya and Basu 1993). The Upper Pleistocene–Early Holocene Toba-ash-bearing unit is mainly represented by coarse alluvium that often contains profuse laterite pisolite as clasts and is associated with calcretes. In places, these sediments are feruginized to form bedded laterites. This Quaternary unit is disconformably succeeded by unoxidized argillaceous alluvium, which also contains calcrete. Based on proximity, lithological and organizational similarity, the Toba-ash-bearing basal Quaternary unit is tentatively correlated with the younger laterite topped unit, which is recognized in the subsurface from the western marginal areas of the Bengal basin. Thus, the laterite plain of West Bengal may be tentatively assigned to Pleistocene or early Holocene age, whereas the so-called OAP in West Bengal is early Holocene in age (Fig. 2).

Similarly, in Bangladesh, the Madhupur and Barind uplands, and the Lalamai hills, which are free from the arsenic problem, are characterized by deeply oxidized palaeosol and associated alluvium that may be broadly contemporaneous (Morgan and McIntire 1959; Alam and others 1990; Monsur 1995). The lower parts, or the entire section of moderately oxidized alluvium, which disconformably overlies these deeply oxidized beds, have yielded reverse magnetic polarity (Monsur 1995) that corresponds to the top of the Matuyama reverse chronology of 0.73 Ma age, whereas, the underlying and overlying formations show normal magnetic polarity. Thus, the Madhupur and Barind Tract sediments broadly correspond to early Pleistocene age (Monsur 1995). The presence of the youngest Toba-ash bed marker (75,000 years b.p.) has been recognized locally over the Barind areas of the Bengal basin (Abdullah and Hasan 1991). These upland terraces from West Bengal and Bangladesh are free of arsenic-toxicity problems.

The OAP in West Bengal is truncated and superimposed by the lower level terraces of YDP, which, in turn, is incised by the present channel and delta plain of the River Bhagirathi. Thus, parts of the OAP fill have been eroded and parts may have been preserved beneath the western cover of YDP. The latter would get mingled with equivalent deltaic sediments that correspond to the Bhagirathi tributary channel further east. Although YDP is definitely younger than OAP, because of an absence of any recognizable corelatable markers, the subsurface equivalents of an OAP morphostratigraphic unit cannot be recognized and delineated beneath YDP. The stratigraphy of Holocene sediments beneath YDP, within which arsenic-bearing aquifers are located, is mainly inferred from limited subsurface data. Local names have been assigned to these subsurface units from area to area. Arsenic-toxic YDP sediments also partly encroach into the Barind area, north of Ganges channel, e.g. the Malda Formation in West Bengal or the Rohanpur/Chalonbill formations of Holocene age in Bangladesh (Monsur 1995). These Holocene valley fills of silt and clay are deposited in deeply eroded and incised Barind sediments (Morgan and McIntire 1959) of Pleistocene age.

Based on generalized lithology inferred from groundwater drilling and heavy mineral studies, the subsurface sediments beneath the YDP from northern and southern arsenic-affected districts in West Bengal have been tentatively classified into three broad stratigraphic zones (Deshmukh and others 1973; Biswas and Roy 1976; Centre for Study of Man and Environment [CSME] unpublished report 1991; Table 1, Fig. 3A). Most arsenicferous tube wells generally tap the aquifers in the stratigraphic zone, Unit II, and the arsenic content in groundwater is generally higher in aquifers in Unit II compared with the other two units. Unit II is dominantly made up of fine sand and clay, whereas Unit I is coarser and sandy. Unit II also outcrops or occurs at very shallow depth in the northern area, e.g. Malda and Murshidabad districts, but regionally dips southward, the dip being steeper than the regional slope. Biswas and Roy (1976), who worked around Raninagar in eastern Murshidabad, recognized a sheet-like gravel-bearing sandy unit from the basal parts of the Quaternary sequence, which disconformably overlies the Upper Tertiary clays (Fig. 3A). These wells are located not far from the present Ganges channel, and the subsurface sheet of sand and gravel may represent the braided channel lag deposit of proto-Ganges. Biswas and Roy (1976) have recorded lenses of calcrete nodules from different levels of the section. This gravel-bearing unit was not reached in many shallow wells. Further, such facies may not have developed uniformly. Similar sand and gravel units have been recognized at sites that occur beneath a clay-silt dominant unit (Fig. 3A). Such subsurface sections are located not far from the present Bhagirathi River course in West Bengal (Deshmukh and others 1973; Hait and others 1996). It was encountered at 170 m depth at Sagar Island. This sand and gravel unit, where developed, possibly represents the basal part of the stratigraphic zone, Unit I. In south-eastern parts of West Bengal, some deep tube wells that tap the aquifer at 100 m or more in depth, i.e. apparently corresponding to Unit I, have also became arsenicferous with continued groundwater extraction, e.g. in the Ghetugachi and other areas in Nadia, and in 24 Parganas districts. Such deeper aquifers are comprised of fine sand interbedded with clay and silt at their lowermost drilled section.

An attempt has been made to correlate the units recognized from the Quaternary section of the Bhagirathi-Ganges delta plain in West Bengal with those of the Brahmaputra-Jamuna braided river floodplain and southern parts of the Ganges delta in Bangladesh (Table 2, Fig. 3B, after Umitsu 1993). Sand with scattered gravels,

Table 1
Raninagar, District Murshidabad, West Bengal, Inland region north. (After Biswas and Roy 1976; CSME unpublished report 1991)

Unit	Depth range (m) b.g.1.	Generalized lithology	Range of arsenic in groundwater (mg/l)
III (not developed or condensed section)	0–6	Clay and sandy clay	0.2–6.0
II	6–50	Fine- to medium-grained sand with clay intercalations	3.2–93.1
I	40–70	Medium-grained sand	2.0–5.2 (not sampled)
	60–80	Pebbly sand	
Disconformity			
Upper Tertiary	+70–+80	Silty clay	(not sampled)
Average arsenic content: Clay −12 ppm, sand −4.8 ppm			
Ashoknagar, Halderbagan, District 24 Parganas (north), West Bengal, Inland region central			
III	0–20	Clay	0.1–6.2
II	20–55	Fine- to medium-grained sand with clay bands	0.1–25.0
	55–70	Clay with fine-grained sand	2.0–15.0
I	70–115	Fine- to medium-grained sand with clay	0.5–5.5
Average arsenic content: Clay 9.5 ppm, sand 3.8 ppm			

with a basal sand-gravel bed of ~10-m thickness, represents the lower unit from the Brahmaputra-Jamuna floodplain. The average gradient of the upper contact of the basal bed is steeper than the gradient of the present braided river plain. The top section of this unit is oxidized in places and its upper contact is usually sharp. The sediments of the middle unit are composed of silt or clay in the lower part and sand in the middle and upper parts. Fine sediments dominate the Ganges delta, especially near the present coastal zone. The lower part of the upper unit consists of clays or silts, even in the inland region, and the upper part of the unit consists of silts and sands with occasional peat layers.

C^{14} dating of fossil wood from the basal part of Unit 1 from the Brahmaputra-Jamuna floodplain, Bangladesh, from an unreported (but apparently northern) location and at 62 m depth, and samples from Sirajganj at 112 m depth (101 m b.s.l.) resulted in dates of 48,500 and 28,300±1500 years b.p., respectively (Umitsu 1993; Anonymous 1999). In comparison, organic matter from the uppermost part of Unit 1 at a depth of 48 m (46 m below sea level [m.b.s.l.]) from the Khulna area, located in the southern parts of the Ganges delta has given a date of 12,300±240 years b.p. At Khulna, the thickness of oxidized sediments at the top of Unit 1 is ~15 m. The top sandy section of Unit I is also oxidized at places in West Bengal (Poddar and others 1993). Clay beds interbedded with gravel and overlying a >6 m medium-grained sand bed from Digha, located at the western boundary of the Bengal basin and on the present coastline (Fig. 1), have yielded a date of 22,360 years b.p. (Hait and others 1996). Further, a similar sand bed with calcretes, silt and clay layers from Daimond Harbour, has yielded 14,460 years b.p. (Hait and others 1996). These beds may be correlated with Unit I.

These ages broadly corroborate the hypothesis that the sand with gravel basal unit, recorded at the Brahmaputra-Jamuna floodplain and those in the Ganges delta, was deposited as valley fill during late Pleistocene and early Holocene under a low stand condition of sea level. Further, the upper unit from Bangladesh and West Bengal aged ~7000 years b.p., and younger in age, are also lithologically very similar and comparable (Tables 1 and 2). Their uppermost section often has peaty layers in southern parts of the Ganges delta in Bangladesh and West Bengal. Marshes with peat development also extended to the Sylhet Basin (Umitsu 1993). The presence of typical estuarine elements, which range from swampy mangrove to tidal mangrove, with an initial *Ammonia* transgression phase in brackish to fresh water sediments, is recorded by Banerjee and Sen (1987) in the Bhagirathi delta, West Bengal. These sediments occur at 7 to 2 m b.s.l. depth and are located in areas 80 to 120 km inward from the present shoreline. Based on C^{14} dating these high sea-level sediments are tentatively dated at 7000 to 6000 years b.p. Sediments found at depths of 2 m b.s.l to s.l., and dated between 6000 and 5000 years b.p., indicate the presence of fresh-water- and mixed-brackish-water-forest. Fresh-water bioassemblage is recorded from sediments dated 5000 years b.p., and is younger in age that occurs at present sea level to +6 m. There was widespread development of marine and fresh-water peat layers around Calcutta, and at ~60 km north, during 6500–2000 years b.p. (Banerjee and Sen 1987). Good aquifers are absent from this unit and arsenic-contaminated aquifers in West Bengal are generally located well below this peat-bearing zone.

Thus, the middle unit, which contains the arsenic-bearing aquifers from West Bengal and Bangladesh (Tables 1 and 2), may also be broadly correlated. However, in the Brah-

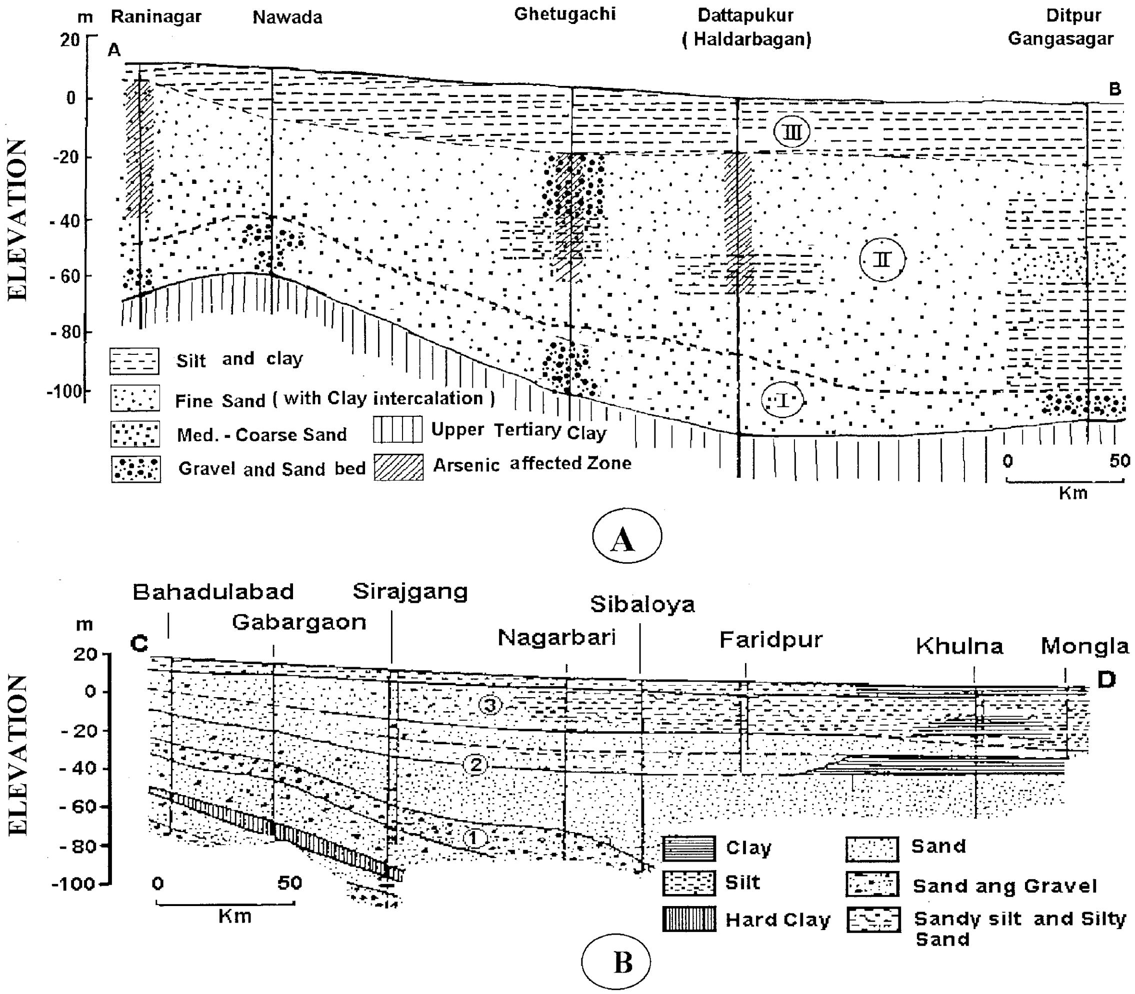

Fig. 3
Profiles of Holocene sediments, lines of section shown in Fig. 1. **A** Section *A–B* across Ganges delta in West Bengal, and **B** *C–D* across Jamuna floodplains and Ganges delta in Bangladesh after Umitsu (1993). *I–III* and *1–3* are broad stratigraphic units

maputra-Jamuna braided river basin that is located inland, equivalent sediments, which are coarser and sandy, are not affected by arsenic toxicity. From C^{14} dating of organic matter from various levels of middle unit and the base of upper unit, the age of Unit 2 has been broadly inferred to be 10,000–7000 years b.p. (Umitsu 1993; Hait and others 1996). From the cores at Khulna, fossil molluscs and marine planktonic diatoms have been recorded at depths of 20–35 m in Unit 2, and they nearly cover the entire unit. This indicates a relatively strong marine influence. Thus, the coastline of the Ganges delta was located to the north of Khulna and Calcutta during the

Table 2
Brahmaputra-Jamuna braided river floodplain and lower Ganges delta plain. (After Umitsu,1993; Anonymous 1999)

Unit	Brahamaputra-Jamuna floodplain, inland region (Tangail) Generalized lithology	Ganges delta, coastal region (Khulna) Generalized lithology	Approx. age 48,500–12,000 years b.p. Based on C^{14} dating from organic matter
3	Silt	Silt with peaty clay	Present
	Silty sand	Peat, peaty silt	~7000 and younger
2	Medium-grained sand	Sandy clay	
	Silty sand	Clay	~10,000–7000
	Break represented by oxidized sediment at top		
1	Sand with scattered gravel	Medium- to fine-grained sand	~48,500–12,000

deposition of the arsenic-bearing middle unit. At about the same time, marshy lands developed inland in the Sylhet Basin (Umitsu 1993), and these appear to have hosted arsenicferous aquifers.

Sedimentary environments and sea-level changes

Late Pleistocene and Holocene sedimentation in the delta plain of the Bengal basin has been strongly influenced by global changes in sea level (Niyogi 1975; Bhatacharya and Banerjee 1979; Banerjee and Sen 1987; Umitsu 1993). It is possible to reconstruct the late Pleistocene–Holocene sedimentation history for the Bengal basin based on the character of sediments, stratigraphy, sea-level changes and landform evolution. There appears to be a positive correlation between the location of arsenic-bearing zones with deltaic environment and organic-rich deposition during the mid-Holocene sea-level rise.

The basal sand and gravel beds were deposited during late Pleistocene–early Holocene in low sea-levels in the Ganges-Brahamaputra delta. The sea level gradually decreased and reached a lowest level of ~135 m during late Pleistocene (~18,000 years b.p.). The shoreline was then located at the outer margin of the continental shelf, and exposed the Pleistocene and late Tertiary sediments to subaerial erosion. Parts of the Pleistocene cover around the present delta region remained as incised upland terraces in the west, north and south-east. In response to lower sea level, the present combined delta and surrounding areas were subjected to valley incision and fluvial sedimentation. The basal sand and gravel unit was not uniformly developed, but was restricted to incised valley courses (Fig. 4). The Ganges-Brahmaputra River possibly drained through the 'Swatch of no ground' to the Bay of Bengal (Poddar and others 1993). Proto-Bhagirathi also existed and spread to the Bay of Bengal around the position of present-day Sagar Island. From 18,000 to 12,000 years b.p., the sea level continued to rise causing transgressional overlapping sedimentation that filled entrenched valleys with fluvial or fluvio-deltaic sand, with some silt and clay. Some of these sediments were also directly deposited over the eroded Pleistocene or Upper Tertiary sediments in the delta plain and surrounding area. From 12,000 to 10,000 years b.p., parts of the Ganges delta appear to have been subjected to weathering and oxidation, possibly caused by temporary regression.

The middle unit was deposited around 10,000 to 7500 years b.p., when the sea level rose continually till 9500 years b.p., to reach ~ −20 m and then broadly maintained this level till 7000 years b.p.(Nelson and Bray 1970). The rising sea level at the initial stage would have flooded the partly sedimented entrenched valley courses of proto-Bhagirathi and the Ganges-Brahmaputra, and converted their lower and adjacent parts to fluvial marshes, lagoons and estuaries (Fig. 4). A swamp would be formed ahead of the rising sea level. High tides in the Bay of Bengal would advance the tidal flat and mangrove growth to invade the delta mouth, which would clog with mud and silt rich in organic content. Thus, clay and silt that are rich in organic matter and interbedded with lenticular sand bodies from numerous transient tributary channels dominated the middle unit. Subsurface data indicate that the shoreline was then located north of Khulna and Calcutta (Fig. 4).

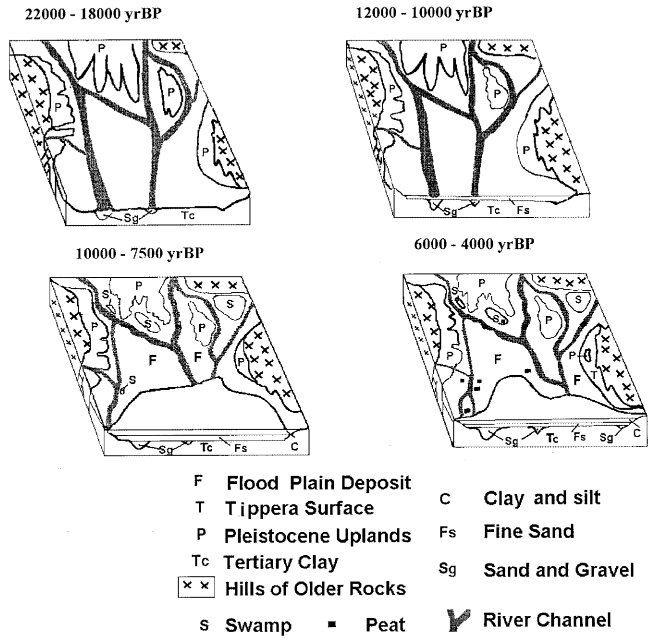

Fig. 4
Palaeogeographic block diagrams of parts of Bengal basin during Holocene. (Modified after Umitsu 1993)

Sea level again began to rise rapidly between 7000 to 5500 years b.p., to reach higher than present-day levels, and southern parts of the Ganges delta were further invaded by tidal mangrove that encroached the Bay of Bengal. There was extensive development of marine and fresh-water peat. After this post-glacial optimum, the sea level dropped initiating a phase of subdued marine regression and migration of the shoreline to the present-day conditions. The depositional environment in the Ganges delta was not much different during the middle and the upper unit. A peaty clay layer at 9 m depth in the Ghetugachi area contains 50 ppm of arsenic. The upper unit, however, usually does not contain good aquifers.

Source of arsenic in groundwater

The wide expanse and regional extent of the arsenic-toxic zone in the Ganges delta in West Bengal and Bangladesh, as well as, those in the Meghna River floodplain and in the Sylhet basin, Bangladesh (Fig. 1), which all have different catchment areas, indicates multiple natural sources of arsenic contamination. In contrast, the Brahmaputra-Jamuna, and old Brahmaputra floodplain in Bangladesh, are so-far unaffected although these areas are also subjected to similar levels of groundwater extraction compared with other areas that are found to be arsenic toxic. A plausible explanation will be discussed later.

The specific source of arsenic contamination in the Ganges basin is as yet unidentified. Likely sources for arsenic in the Ganges delta may include the following (Fig. 1): (1) the Gondwana coal seams in Rajmahal basin contain up to 200 ppm of arsenic (the overlying basaltic volcanics are not enriched in arsenic), (2) the arsenic mineral lollingite and pyrite, which occur sporadically in association with pegmatites in the mica-belt of Bihar, have an arsenic content in mineralized rocks that ranges from 0.12–0.018% (Bhattacharya 1972), (3) pyrite-bearing shale from the Proterozoic Vindhyan range, with its Amjhore mine, contains 0.26% As (Das 1977), (4) the gold belt of the Son Valley has an arsenic content in the bedrock that locally reaches 2.8% to 1000 ppm (Mishra and others 1996), (5) isolated outcrops of sulphides contain up to 0.8% arsenic in the Darjeeling Himalayas, and (6) in outcrops in the upper reaches of the Ganges River system. The copper belt of Bihar, and the Damodar valley Gondwana coal basins contain local concentrations of arsenic in pockets that are drained by rivers that flow far to the south of the Ganges tributary system, and thus these cannot be potential sources as postulated by some (Nickson and others 1998; Acharyya and others 1999). Further, the Damodar alluvial fan that flanks the arsenic-affected Bhagirathi delta is free of arsenic problems.

The Sylhet basin is flanked to the north by the Proterozoic and Tertiary shelf sequences, which cover the top and southern parts of the Shillong hills, respectively. Local sulphide mineralization in the former occurs near the water divide and contains pyrite, arsenopyrite and lollingite (GSI, unpublished report). The Eocene sediments contain coal, sandstone, shale and limestone and these are often pyrite-bearing. Samples from these pyritic sandstone and shale often contain 10–50 ppm arsenic, and the limestone locally contains up to 154 ppm arsenic. The younger Tertiary sediments also contain some pyritic sediments. Thus, drainage from the Shillong hills is likely to carry arsenic to the Sylhet basin. Drainage from the eastern part of the Shillong hills also feeds the floodplains of the Meghna River (Fig. 2). Further, the catchment area of the Meghna River includes Tertiary sediments, some of which have pyrites. Thus, on weathering, these may be a potential source of arsenic. The Brahmaputra-Jamuna and old Brahmaputra floodplains are free of the arsenic problem. The upper reaches of Brahmaputra, before entering the Bengal basin, are fed by Himalayan streams and those draining the basement gneisses of the northern face of the Shillong Plateau (Fig. 1). No specific potential source of arsenic is as yet known from this catchment area.

The ongoing mineralogical studies indicate that arsenic-rich pyrite and separate arsenic minerals are absent or rare in the sediments of the Ganges delta from West Bengal. However, arsenic appears to be adsorbed to iron-hydroxide-coated sand grain margins and to clay minerals (Acharyya and others 1999). The rare presence of authigenic pyrite has been recognized from the Ghetughchi area and is also reported in aquifers from Bangladesh (Nickson and others 1998; Anonymous 1999). In the Ghetughachi area, the presence of arsenic is noted in a rare case within iron hydroxide- and sulphide-coatings on heavy mineral illmenite. Thus, it is likely that arsenic was co-precipitated with or scavenged by iron(III) and manganese(IV) in the sedimentary environment. Thus, the presence of arsenopyrite or the pyrite-bearing layers or zones in the aquifer, as has been postulated by some (Das and others 1996; Mallick and Rajagopal 1996) is doubtful. Instead it is likely that decomposition of pyrite and arsenic-bearing sediments in the source area released arsenic in a soluble form, and this was sorbed by secondary iron oxyhydroxide. Arsenic in solution possibly became more easily entrapped in the fine grained organic-matter-rich sediments that were deposited preferentially under low energy conditions in the Ganges delta, Meghna floodplains and the Sylhet basin during the mid-Holocene sea-level rise. Sediment thickness in these domains was also appreciable because of tectonic subsidence during sedimentation. However, the alluvial tract of the Ganges upstream of the Rajmahal hills in Bihar and Uttar Pradesh (UP), which is joined by numerous tributaries from the Himalayas, is sandier and much thinner compared with the delta region. This may have contributed to lower initial retention of arsenic in this section. The catchment areas of the Brahmaputra-Jamuna River are not known to contain potential sources of arsenic and, further, the relatively coarse sediment that fills these braided streams is not efficient at retaining arsenic.

Mobilization of arsenic in groundwater

Organic-rich sediments from the Ganges delta, the Meghna floodplain and the Sylhet basin would create strongly reducing conditions. This process would increase with depth and with the high water table and clayey surface sediment layers that impede entry of air to the adjacent aquifer. The highly reducing nature of the groundwater would reduce As to As(III) and cause the possible desorption of As, because As(III) is normally less strongly sorbed by iron oxide than As(V) under near-neutral pH. Further reduction will lead to partial dissolution of poorly crystallized ferric oxide with the consequent release of iron and additional As. Dissolved iron content in groundwater is a measure of the degree of reduction. The dissolved iron in groundwater in the Ganges basin in UP and the Bihar states in India is reported to have trace concentrations of 1.0 mg/l compared with values of up to 36 mg/l in the south of West Bengal (Acharyya and others 1999) and 30 mg/l in Bangladesh (Nickson and others 1998). The relatively low values of dissolved iron upstream of the Ganges floodplain in Bihar and UP possibly indicate that the environment is not sufficiently reducing to mobilize iron and arsenic. Iron concentration in groundwater from Brahmaputra-Jamuna and the old Brahmaputra floodplain in Bangladesh ranges from 8.5 to 9.0 mg/l (Anonymous 1999; Table 2), which indicates a prevailing reducing environment, but here the sediments themselves appear to be deficient in arsenic content. The reduction process is certainly mediated by microbial action. The recent arsenic contamination in groundwater during the last three decades in the Ganges delta and other regions in Bangladesh appears to be linked to excessive groundwater extraction for irrigation. Since the1960s, over 0.5 million tube wells with hand-pumps, about 0.1 million shallow tube wells, and about 3000 deep tube wells were sunk in Gangetic West Bengal, at depth ranges of 10–20, 30–100 and 50–200 m, and with low, 20 and 100 m^3/h average discharges, respectively (Bagla and Kaiser 1996; Mallick and Rajagopal 1996). Similarly, in Bangladesh, groundwater irrigation began in the mid-1970s. It has increased rapidly since the1980s and had reached over 3 million ha in 1995 (Anonymous 1999). Simultaneously, the application of phosphate fertilizers has tripled because of switching from one crop to three crops per year. It is possible that phosphate derived from fertilizers, as well as from the decay of natural organic matter, has been mobilized into the aquifers because of widespread irrigation and groundwater withdrawal. It is well known that phosphate encourages growth of sediment biota. In addition, phosphate may also act as an eluant for arsenic from sediment. Preliminary experiments, similar to the extraction method of Manning and Martens (1997), indicate that 1 mM KH_2PO_4 solution can leach out up to 150 μg/g total As from Fe-coated quartz grains in sandy aquifers of the Ghetugachi area. These combined microbiological and chemical processes might better explain the recent release of arsenic in groundwater and the general trend of decreasing arsenic concentration with depth (Acharyya and others 1999).

Conclusion

Arsenic toxicity in groundwater affects major parts of the Bengal basin, such as the delta plains of the Ganges downstream of the Rajmahal hills and some low-lying areas located in the eastern and southern parts of the basin. The affected sediments in the area and stratigraphic level are confined to fine grained and organic-rich low energy sedimentation in the delta area during mid-Holocene sea-level rise. Dissected Pleistocene and early Holocene terraces and highlands that occur within and flank the Bengal basin are unaffected. Some potential natural but lean sources of arsenic have been identified. The presence of arsenic mineral has not been recognized in the affected sediments and arsenic appears to be adsorbed on iron hydroxide-coated sand grains and clay minerals. Arsenic transported in solution was sorbed by iron oxyhydroxide and became preferentially entrapped in deltaic and marshy organic-rich sediments. It was liberated subsequently by microbially-mediated reduction reactions in such sediments. The environment is not sufficiently reducing in the Ganges floodplains upstream of Rajmahal to mobilize iron and arsenic in groundwater. However, Jamuna and the old Brahmaputra River floodplains in Bengal basin are free of arsenic because of the absence of a potential source and because of a coarser sediment fill, despite reducing conditions sufficient to mobilize iron in groundwater. The recent release of arsenic in groundwater in the Ganges delta and from some low-lying areas in the Bengal basin is possibly triggered by intensive groundwater extraction for irrigation and the application of phosphate fertilizer. This has induced movement of groundwater, and the mobilization of phosphate derived from fertilizer, as well as from decayed organic matter. This would, in turn, promote growth of sediment biota and aide in the further release of arsenic. Preliminary experiments indicate that phosphate solution can leach out significant amounts of arsenic from Fe-coated quartz grains in sandy aquifers of the Ghetugachi area, West Bengal. Thus, arsenic toxicity in groundwater in Bengal basin is caused by its natural setting, but appears to be triggered by recent anthropogenic activities.

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