



Sources of low-arsenic groundwater in the Bengal Basin: investigating the influence of the last glacial maximum palaeosol using a 115-km traverse across Bangladesh

M. A. Hoque · J. M. McArthur · P. K. Sikdar

Abstract Pollution of groundwater in the Bengal Basin (Bangladesh and West Bengal, India) by arsenic (As) puts at risk the health of more than 100 million consumers. Using 1,580 borehole lithological logs and published hydrochemistry on 2,387 wells, it was predicted that low-As (<10 µg/L) groundwater exists, in palaeo-interfluvial aquifers of brown sand capped by a protective palaeosol, beneath at least 45,000 km² of the Bengal Basin. The aquifers were predicted to be at a depth of as little as 25 m below ground level (mbgl), and typically no more than 50 mbgl. The predictions were confirmed along an east–west traverse 115 km in length (i.e. across half of Bangladesh) by drilling 28 new boreholes to 91-m depth to reveal subsurface sedimentology, and by mapping As distribution in groundwater. The aquifers identified occur at typically <40 mbgl and so are accessible with local drilling methods. A protective palaeosol that caps the palaeo-interfluvial aquifers prevents downward movement into them of As-polluted groundwater present in shallower

palaeo-channel aquifers and ensures that the palaeo-interfluvial aquifers will yield low-As groundwater for the foreseeable future. Their use, in place of the shallower As-polluted palaeo-channel aquifers, would rapidly mitigate the health risks from consumption of As-polluted groundwater.

Keywords Arsenic · Contamination · Bangladesh · Palaeo-interfluvial · Palaeo-channel

Introduction

In Bangladesh and West Bengal (India), i.e. the Bengal Basin, at least 85 % of the combined populations of 240 million use groundwater for domestic water supply, including for drinking. The groundwater is widely polluted with arsenic (PHED 1991; DPHE 1999, 2001; van Geen et al. 2003b; Jakariya et al. 2007; Nickson et al. 2007; Fendorf et al. 2010), the long-term consumption of which degrades human health (Dhar et al. 1997; Smith et al. 2000; Argos et al. 2010). Mitigation strategies include rainwater harvesting, use of treated surface-water supplies, and exploitation of deep aquifers (>150 m below ground level (mbgl) and mostly >300 mbgl) which typically contain much less than 10 µg/L of As (DPHE 1999, 2001; Ahmed et al. 2006; van Geen et al. 2007; Ravenscroft et al. 2009). These strategies require capital investment and maintenance that is beyond the capacity of most of the population, given that 90 % of it lives in a rural environment sustained by subsistence farming.

Cheaper mitigation can be obtained by emplacing well screens in aquifers of brown sand, where they are found in the subsurface, typically at depths between 25 and 70 mbgl. Recognised as distinct aquifers by Davies and Exley (1992) and Davies (1995), the brown sand's connection to low-As groundwater was established by P. Ravenscroft of the Department of Public Health Engineering (DPHE) of Government of Bangladesh (DPHE 1999), and confirmed by others (DPHE 2001; van Geen et al. 2003a; McArthur et al. 2004; von Brömssen et al. 2007). Such aquifers contain groundwater in which concentrations of As are <5 µg/L and not uncommonly <1 µg/L. Other work by Goodbred and

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Kuehl (1999, 2000) suggested that the presence of such brown sands, as part of their ‘oxidised facies’, may be widespread across the Bengal Basin, although those authors did not link them to low-As groundwater. With hindsight, the development of the Bengal Basin outlined by Umitsu (1993) also allows the inference to be drawn that palaeo-interfluves are widespread across the basin and so, by further inference, are brown-sand aquifers.

Mitigation by exploiting aquifers of brown sand is an approach that is at risk from migration downward of As-polluted groundwater in overlying aquifers. The risk is removed where the brown-sand aquifers are protected from downward recharge by a capping red-clay palaeosol, first noted in central Bangladesh (Davies 1995), that formed much of the land surface (i.e. palaeo-interfluves) during the last glacial maximum (Goodbred and Kuehl 1999, 2000; McArthur et al. 2004, 2008). The combination of capping palaeosol over brown sand is seen most prominently as the upstanding Madhupur and Barind Tracts. The subcrop of this palaeosol represents the buried palaeo-interfluvial regions of the Bengal Basin. If truly widespread and not subsequently removed by erosion, those palaeo-interfluves, now known to host brown-sand aquifers, would constitute a substantial source of low-As groundwater that could be used to mitigate As-pollution in the Bengal Basin. Such a strategy might be applicable to other low-latitude deltas where groundwater is exploited for human consumption, e.g. the aquifers of the Indus (Pakistan), Red and Mekong (Vietnam), Pearl and Yangtze (China) river deltas.

Given the potential importance of these palaeo-interfluvial aquifers (brown sand capped by a palaeosol) to the provision of low-As water across Asian deltas, it is important that their distribution be documented, and their true value as sources of low-As water is assessed. A start to such explicit mapping and assessment has been made in the Bengal Basin by McArthur et al. (2008, 2011, 2012) and Hoque et al. (2012), who document areas of palaeo-interfluvial aquifer across parts of West Bengal.

The work reported here has extended that mapping across a large part of Bangladesh by using a combination of groundwater composition and historic drilling logs to distinguish, in the subsurface, between aquifers in palaeo-channel (PC) settings that host As-polluted groundwater, and aquifers in palaeo-interfluvial (PI) settings that host low-As groundwater. These methods suggested that largely unexploited PI aquifers yielding low-As groundwater exist beneath about one third of Bangladesh and also suggested that they are common in areas where presently almost all groundwater exploitation is from shallower, As-polluted, PC aquifers. Those suggestions were tested by drilling 28 new boreholes along a 115-km traverse across Bangladesh, by doing further analysis for chemical proxies in groundwater of PI/PC settings, and by undertaking a new colour survey of wells (McArthur et al. 2011) that enables PI/PC settings to be distinguished in the subsurface without drilling or chemical analysis of groundwater.

Study area and its sedimentological development

The study area is shown in Fig. 1 and comprises much of southern and central Bangladesh. The exploration traverse (Fig. 1) extends due east from the border between Bangladesh and India and has its eastern end within 20 km of the Ganges (locally termed Padma) River. This traverse crosses much of the As-polluted region of Bangladesh.

The climate of the region is monsoonal, and delivers between 1.5 and 3 m of rain per year, with the amount increasing from SW to NE. The rain occurs mainly between May and October (Sengupta and Sarkar 2006). The water table is typically only a few metres below ground level but deepens a few metres during the dry season (November–April) in response to natural drainage to rivers and abstraction for domestic use and irrigation. The water level locally may go temporarily below the level at which it can be raised by the suction pumps used for irrigation and domestic supply (typically <7.5 m/25 feet). The shallow aquifers across the Bengal Basin (i.e. <150 m depth) are fully recharged each year by monsoonal rain, with the water table rising above ground level in many areas during the monsoon to cause local inundation of lower ground with rainwater.

The present Bengal Basin comprises numerous major and minor river channels and the extensive interfluves that lie between them. A similar landscape would have existed around 125 ka, when sea level was around the level that it is today. In the interval 125 ka to the last glacial maximum at 18 ka (LGM), sea level fell by 120 m. In that period, weathering and erosion developed a late Pleistocene landscape at an elevation mostly lower than that of today composed of interfluves and river valleys (Umitsu 1993; DPHE 1999; Goodbred and Kuehl 1999, 2000), the most prominent of which remain today as the Barind and Madhupur Tracts. Long weathering of the palaeo-interfluves formed on them a capping laterite of red clay (Goodbred and Kuehl 1999, 2000; termed here the last glacial maximum palaeosol, or LGMP) over weathered and oxidised sands that are now commonly termed ‘brown sands’ (DPHE 1999 *et seq.*) and represent part of the ‘oxidised facies’ of Goodbred and Kuehl (2000).

The rise of sea level after the termination of the LGM created the accommodation space for the accumulation of newer sediment on top of the late Pleistocene landscape. First, the pre-LGM palaeo-channels, incised to a maximum depth of 120 m, were filled with mostly grey sands. Later sediments consisted of post-LGM floodplain silts, peats, and clays, incised by numerous later channel-fills of grey sands deposited as a result of channel avulsion. The channel-fills range in formational age from immediate post-LGM, in the base of main distributary channels, to modern deposits arising from modern river avulsion (DPHE 1999; Goodbred and Kuehl 1999, 2000; chapter 3, Ravenscroft 2003).

Within this sedimentological context, four sedimentological settings are recognised here (Fig. 1b) which have general meaning for As pollution. The channels present at

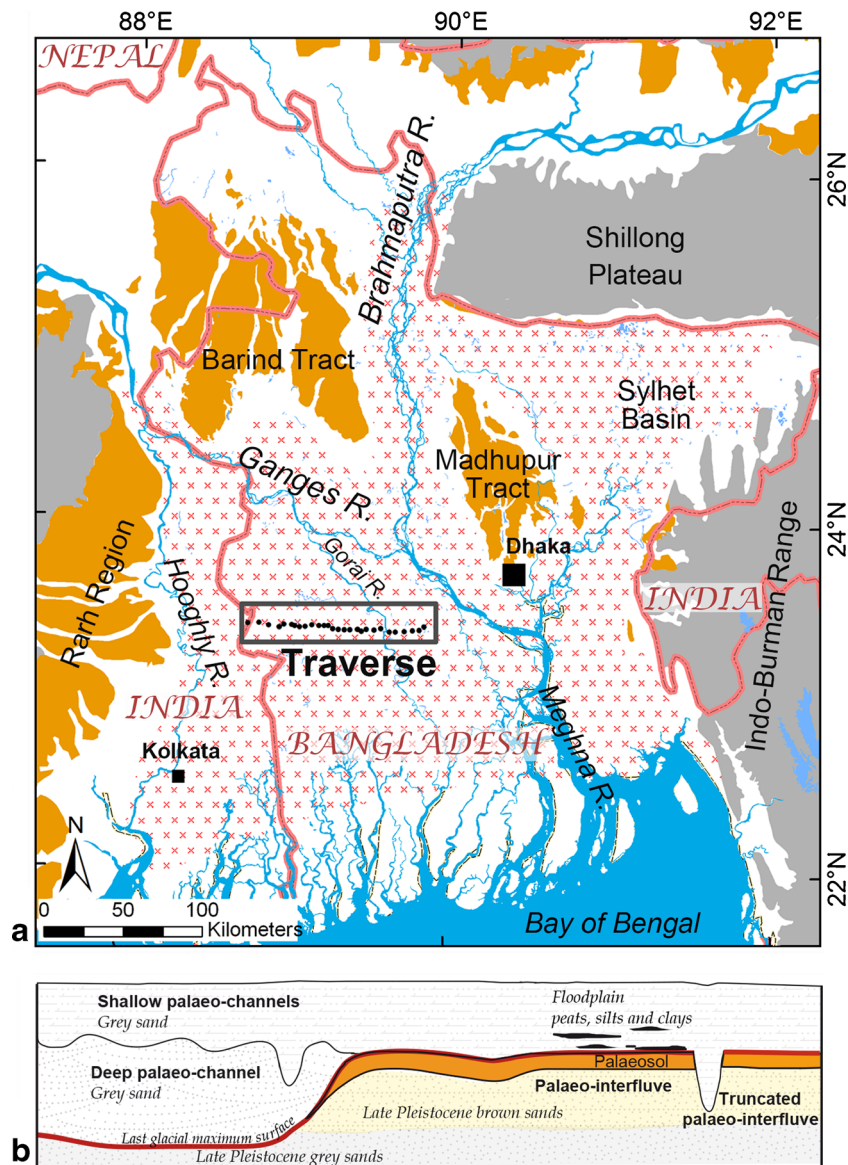


Fig. 1 a Map of Bengal Basin, showing regions most affected by As-polluted groundwater (red cross-hatch). Boxed is the traverse of 28 boreholes drilled to reveal subsurface sedimentology. Around each borehole, field analysis for As in water wells, and well-colour surveys (see section ‘Methods’ for details) confirmed the sedimentology revealed by drilling. Areas of solid dark brown show major outcrops of Pleistocene and older sediments (Madhupur Tract, Barind Tract, Rarh Region), the subsurface extensions of which this work sought to document. b A generalised illustration of aquifer settings (not to scale), showing four aquifer settings described in the text: palaeo-interfluvial, truncated palaeo-interfluvial, deep palaeo-channel, shallow palaeo-channel

the LGM, and infilled with grey sand as sea level rose, are termed here deep palaeo-channels (DPC). Shallow palaeo-channels (SPC), typically <30 mbgl, contain grey sands which were formed by avulsion of Holocene rivers after most of the basin-fill had been deposited. Where the LGMP directly caps late Pleistocene brown sands and underlying Pleistocene grey sands, the term PI setting is used. The term truncated palaeo-interfluvial sequences (TPI) is used for the setting where shallow palaeo-channels have incised through the LGMP, depositing grey Holocene-to-modern sands directly onto brown, late Pleistocene sands.

The SPC aquifers are commonly exploited for water supply because they are the first aquifers to be

encountered during drilling, usually at depths <30 mbgl. The grey sands of the SPC and DPC aquifers usually contain As-polluted groundwater because reduction of sedimentary iron oxyhydroxides, that sorb As, in the aquifer sediments has commonly gone to completion and released that As to groundwater (Gulens et al. 1979; Nickson et al. 1998; Welch et al. 2000; McArthur et al. 2004). Groundwaters in PI aquifers typically have concentrations of As that are <5 µg/L and commonly <1 µg/L because the environment is poised at the redox state of Mn-reduction, rather than the As-polluting stage of Fe-reduction (McArthur et al. 2011).

Brown PI sands and grey PC sands are in contact in TPI sequences and at the margins of palaeo-channels. In

both environments, As-polluted groundwater in the PC sands can invade the brown sands of the PI aquifers. The brown sands have a higher Fe(III)/Fe(II) than grey sands (Horneman et al. 2004) because the brown sands contain more sedimentary iron oxyhydroxide. That iron oxyhydroxide strongly sorbs As (e.g. Stollenwerk et al. 2007). At such contacts, the concentrations of As in PI groundwaters are locally >5 $\mu\text{g/L}$ because the sorption capacity of the brown sands is overwhelmed by As in the invading groundwater (McArthur et al. 2010).

Methods

To map the distribution of PI aquifers across Bangladesh, two methods were used. Firstly, the PI distribution was established using 1,580 rotary-drill logs made available by the DPHE, Government of Bangladesh DPHE/DFID/JICA (2006). Secondly, As, Fe, and Mn concentration of 2,387 well waters from the National Survey of Arsenic Contamination (DPHE 1999, 2001) were used as proxies for PI and PC settings (see the following section ‘Regional water composition’). All locations are reported here in the WGS 84 co-ordinate system.

The distribution of PI aquifers predicted by these methods were then tested by site investigation at 28 locations along an east–west traverse across 115 km of Bangladesh (Fig. 1). At each site, a borehole up to 91 mbgl was drilled to reveal subsurface sedimentology but without installing a well. Also, from existing domestic wells located around each drill site, groundwater samples were collected for chemical analysis of PI/PC proxies. Finally, for a few hundred metres around each site, the colour of stain on further well-completions and on domestic utensils was documented. This was done because such colour can distinguish between PI and PC settings of the well (McArthur et al. 2011) and help locate the boundary between PI and PC settings to an accuracy of a few metres (McArthur et al. 2011; Hoque et al. 2012).

DPHE rotary drilling logs

Using 1,580 rotary borehole logs from known locations that were drilled to depths of at least 100 mbgl, a setting that was either PI or PC was assigned to each log on the basis of the similarity of the recorded lithology to type examples of PI and PC sequences from existing lithological records (Fig. 2; McArthur et al. 2004, 2008, 2011; Hoque et al. 2012). Core descriptions of others were also classified as PI or PC (BADCO/MMI/HTS 1992; Davies and Exley 1992; Umitsu 1993; Goodbred and Kuehl 1999, 2000; Pal et al. 2002; van Geen et al. 2003b, 2007; von Brömssen et al. 2007; Pal and Mukherjee 2009; Burgess et al. 2010; Hoque et al. 2011; Pate et al. 2009), as few had recognised this distinction.

In none of the 1,580 DPHE logs was the distinction between PI and PC sequences recognised previously. It is the re-interpretation of these logs presented here that has

allowed this distinction to be made. The key indicator of a PC sequence is the presence of sand at all depths, usually capped by surficial silt and/or clay typically only a few metres thick. The indicator of a PI sequence in the DPHE logs is the presence, at depths ranging from 26 to 34 mbgl depending on location, of a unit of clay+silt which, where present, was taken to be the LGMP. This is the depth range at which it has either been recognised and reported as such, or reported but not recognised as the LGMP and so re-interpreted here as such (see preceding citations). Further necessary conditions for recognition of the LGMP was a thickness >80 cm and that it was underlain by sands to the maximum depth examined. Where the LGMP was overlain by shallow palaeo-channel sands, identification of the clay between 26 and 34 mbgl as the LGMP is unequivocal as it was the only fine-grained unit reported. Where the putative LGMP is overlain by a sequence of floodplain clays, silts, and peats, the presence of the LGMP was inferred because all such sequences so far drilled by the authors have the LGMP at the base of the clay sequence, under which was a thick sand (McArthur et al. 2001, 2004, 2008; Pal et al. 2002).

The process of assignment was assisted by converting logs to a digital format and processing them using an algorithm that measured the proportions of sand, and silt+clay, present in successive 2 m-thick segments of core. The output of grain-size with depth was diagnostic of either a PI or a PC sequence. The number of logs that satisfy the algorithm was sensitive to the thickness set for that depth slice. That sensitivity analysis suggests that the results must be taken as reporting a minimum number of locations at which a PI sequence may be present. The analysis further under-reports the frequency of PI sequences because the DPHE drilling logs did not record sediment colour. The algorithm therefore classified as PC sequences all TPI sequences, in which grey sands directly overlie brown sands with no intervening LGMP, e.g. the sequences at piezometer site AP in the village of Moyna, West Bengal (McArthur et al. 2004).

A further bias towards under-reporting of the PI sequence occurs because rotary drilling seldom allows recognition of clay/silt units that are thinner than about 3 m in thickness, so the absence of such a unit from a rotary drill log does not indicate with certainty the absence of the LGMP. Despite these caveats, the analysis outlined in the preceding is useful in allowing quantification of the minimum extent of the PI sequences present at modest depth in the Bengal Basin.

Regional water composition

The hydrochemical search for PI aquifers examined water compositions from 2,387 of the 3,234 published analyses of groundwater available from the National Survey of Arsenic Contamination, of Bangladesh conducted in 1997/1998 (DPHE 1999, 2001). Following McArthur et al. (2011, 2012) and Hoque et al. (2012), concentrations of As, Fe and Mn in groundwater were used to differentiate between wells tapping PI sequences and those tapping PC

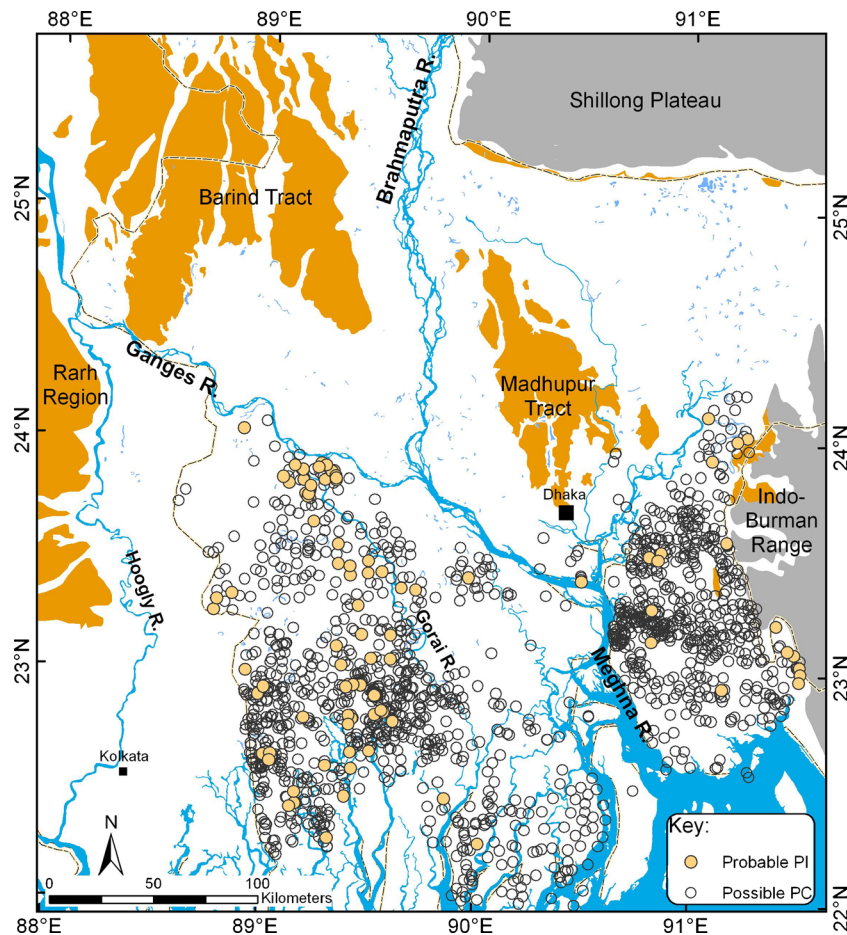


Fig. 2 Locations and classification of 1,580 lithological records used in this study. Sequence categorization is described in the ‘Methods’ section; for reasons given in the text, the proportion of PI settings is underestimated. Lithological logs were supplied by the Department of Public Health Engineering, DPHE, Government of Bangladesh. *Open circles* are locations where lithological logs suggest the presence of PC settings; *filled circles* are locations where lithological logs suggest the presence of PI settings. Areas of dark brown as in Fig. 1

sequences: PI groundwaters typically contain Mn, but no As or Fe; PC groundwaters typically contain As and Fe, but no Mn. Specifically, for wells between 20 and 90 m deep, the criteria set to identify PI groundwaters were: $Mn \geq 0.2$ mg/L and $Fe \leq 0.9$ mg/L and $As \leq 10$ μ g/L. For recognition of PC groundwater, criteria were: $Mn \leq 0.2$ mg/L and $Fe \geq 0.9$ mg/L and $As \geq 10$ μ g/L.

Excluded from consideration were wells screened at depths >90 or <20 m. Brown sands in the Bengal Basin have rarely been reported from depths >90 m, so wells screened below that depth are of limited usefulness in the search for PI brown sands. The shallow cut-off of ≤ 20 m depth is chosen because such wells must be screened in PC aquifers, as the minimum depth at which the PI brown sands have been found in the basin proper is around 25 mbgl. The top of the PI sequence trends to shallower depths as it approaches outcrop around the basin margin, and where it is close to the Barind and Madhupur Tracts, so the upper depth limit breaks down in such settings. Nevertheless, such settings form an insignificant part of the region studied and any anomalies they produce are unlikely to have a noticeable effect on the results or conclusions.

Site investigation

Drilling

The extent of the PI sequences predicted by the two aforementioned methods were tested by drilling 28 boreholes up to 300 foot (91 m) depth along a 115 km traverse running from west to east across half of Bangladesh from its western border (Fig. 1). The traverse crossed much of the As-polluted part of Bangladesh. The boreholes were variably spaced about 3–6 km apart. The local hand-operated reverse-circulation method (Ali 2003) was used for drilling with plastic pipe, which can drill to 300 feet (91 m) rather than the 200 feet (61 m) attainable with iron pipe used in West Bengal. Water wells were not installed in the boreholes. Rather, at each borehole, cuttings were continuously collected and monitored, and were logged every 5 feet (1.52 m) or where a distinct lithological change occurred (see the electronic supplementary material (ESM), in which it should be noted that each 5 foot interval on the photographic logs gives an example only of the sediment(s) recovered in that 5 foot interval). The hand-operated method faithfully recovers clay units (see figure S2, ESM), and can distinguish fine

sand from coarse sands, and both from silts. Comparison of the hand-operated method with percussion coring at the same site (J.M. McArthur, University College London, unpublished data, 2012) had shown that, at all depths, the lithological records were comparable, although sharp lithological contacts in sands, e.g. between brown and grey sands that occur over millimetres in sediment cores, are smeared over a few feet in hand-operated holes.

Water composition

To test the match between sedimentology revealed by drilling, and that determined from groundwater composition, 176 unfiltered samples of well water were collected for laboratory analysis (see table S1, *ESM*) from domestic wells along the traverse, all but two screened at depth ≤ 90 mbgl. The wells were sited within a few hundred metres of the drill sites. Samples were not filtered because it has been shown that the As concentration of unfiltered and filtered waters are indistinguishable (Swartz et al. 2004; Zheng et al. 2005) and the major cation composition of filtered and unfiltered well waters have also proven indistinguishable unless the water is visibly turbid (J.M. McArthur, University College London, unpublished data, 2004); none of the samples were turbid. Samples analysed for U, V, Mo, Mn, Fe, and As were acidified in the field with 0.15 ml of 50 % AnalaR nitric acid. Analysis for Fe and Mn was by ICP-AES; analyses for As, Mo, U, and V, were done using a Bruker 90 ICP-MS.

A further 129 wells (see table S2, *ESM*) were tested in the field for concentrations of As using a digital Arsenator, because in wells between 20 and 90 m deep, the absence of As typically indicates that the well taps a PI sequence. At another 74 sites (see table S3, *ESM*), distributed mostly between drill sites, this field analysis was augmented by noting the colour of stain left by well water on well-completions and domestic utensils. This use of colour follows the demonstration (McArthur et al. 2011) that red stain is typical of wells <150 m deep that tap As-polluted PC groundwaters, whilst black stain is typical of wells that tap low-As groundwater from PI aquifers.

Results

DPHE rotary drilling logs

The DPHE lithological logs that were categorised as PI or PC sequence are plotted on a map of the study area shown in Fig. 2. It is re-emphasised that the number of logs interpreted as representing a PI sequence is underestimated by the analysis of these DPHE logs presented here, so the distribution shown should be viewed as representing the minimum number of such sites.

Regional water composition

The classification of 2,387 groundwater compositions into PC and PI settings are shown in Fig. 3. The putative occurrence of PI aquifers, shown as brown triangles, bears

some correspondence to the region of As-free aquifers at depth predicted in figure 6 of McArthur et al. (2011). Note particularly the high density of wells that scatter SSE from the Barind Tract and reflect the presence of PI sequences at depth.

In Fig. 4 the sites of ‘probable PI’ based on water composition (Fig. 3) are lumped together with those based on lithological logs or drilling (Fig. 2) plus locations of brown sand reported in the literature. The figure shows (in light brown colouration) the predicted areas under which a PI sequence is probably present at depth in the Bengal Basin.

Site investigation

Drilling

The lithological logs obtained along the drilling traverse are compiled in Fig. 5. Examples of the sediments recovered and used to compile the logs are shown in the *ESM*. At 12 sites (5, 7, 9, 11–14, 17, 18, 20, 23, 24), a full PI sequence was found comprising a palaeosol of red-clay, the LGMP, capping brown aquifer sands. At sites 18 and 20, the PI sequence is overlain by fine, floodplain silts and clays; elsewhere, it is overlain by shallow PC sands. At a further four sites (8, 15, 19, 22), the PI and PC sands are separated by a pale-blue clay 1.5–3 m thick, rather than the LGMP, as has been noted to occur elsewhere (Hoque et al. 2012). The pale-blue clay was, in turn, overlain by organic-rich clay of <1 m thick. At a further three sites (6, 10, 21), a truncated PI sequence was found in which post-LGM channel erosion had cut through the palaeosol and overlying strata, but had not eroded much of the brown-sand sequence beneath, leaving grey, As-polluted, PC sands directly on brown PI sands, as has been found elsewhere (McArthur et al. 2011; Hoque et al. 2012).

The depth to the top of the LGMP ranged between 29 and 37 mbgl and the thickness of the LGMP ranged from 0.5 to 5 m. The brown sands beneath the LGMP yielded groundwater containing <0.4 $\mu\text{g/L}$ (median value, $n=77$) of As. The minimum depth to the top of the brown sand was 29 mbgl and the thickness of the brown sands ranged from 4.6 to >55 m. Brown sand extended to the maximum depth drilled (91.4 m) at sites 22, 23, and 24. In other sites where brown sand was found, it passed downward into grey sand at depths between 55 and 86 mbgl. This basal colour transition marks the maximum depth of late Pleistocene weathering, and so approximate palaeo-base level, in past times (Hoque et al. 2012). These depths have not been corrected for subsidence that has occurred since deposition because local subsidence rates are unknown.

At nine sites (1–4, 16, 25–28), grey PC sands were found extending to the full depth drilled. At the ends of the traverse (sites 1–4 and 25–28), the grey sands infill deep and wide pre-LGM palaeo-channels. At site 16, grey sands to the depth drilled infill a channel of width 3–5 km that probably was a distributary river at the time of the LGM. This site may document an early equivalent of the modern-day Gorai River, which is located nearby (Fig. 1).

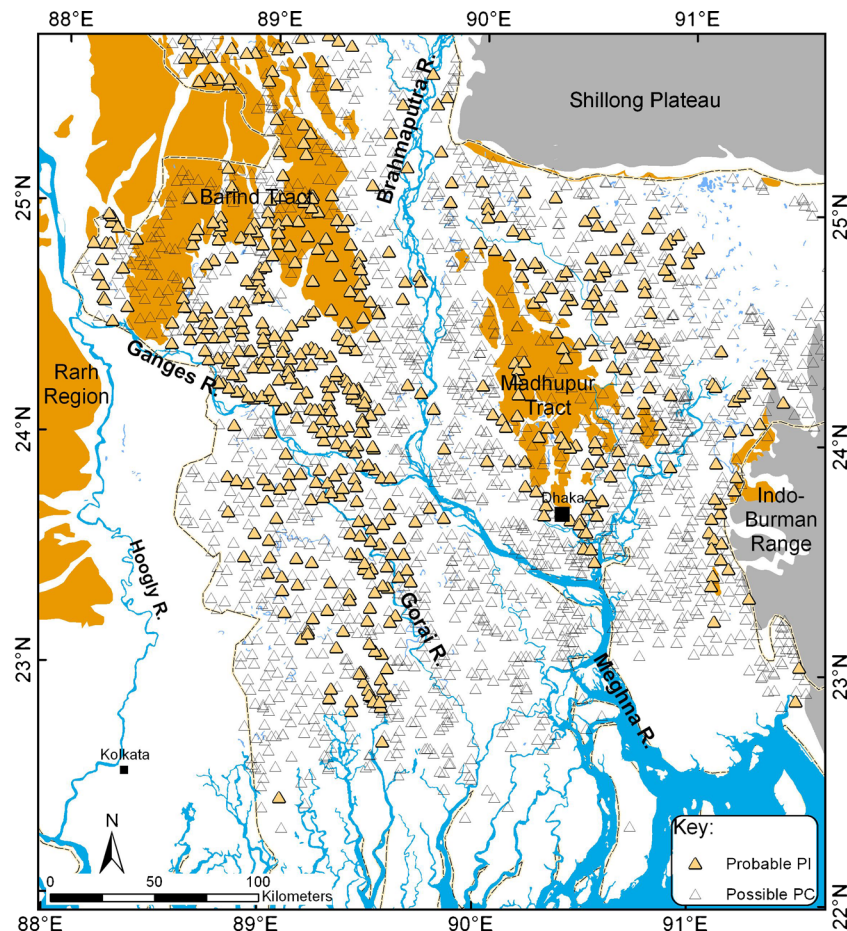


Fig. 3 Locations and classification of the composition of 2,387 well waters of DPHE used in this study. Classification was based on concentrations of Fe, Mn and As in groundwater (see section ‘Methods’). Data from DPHE (1999), (2001). *Open symbols* represent locations where the composition of well water suggests a PC setting; *filled symbols* represent locations where the composition of well water suggests a PI setting. Areas of *dark brown* as in Fig. 1. Note that only wells in Bangladesh were reported in DPHE (1999, 2001)

The shallow PC sands overlying the buried PI sequences typically host As-polluted groundwater (McArthur et al. 2004), but of 14 samples collected in such shallow settings along the line of traverse, concentrations of As exceeded 57 $\mu\text{g/L}$ in one outlier only of 384 $\mu\text{g/L}$. Within the main PI tract, two narrow palaeo-channels occur.

Water composition

The results of well waters collected along the line of traverse are given (see table S1, [ESM](#)). Cross plots of element concentrations, given in Fig. 6, highlight the mutual exclusion of elements in PI and PC groundwater and lend confidence to the use of these element concentrations to separate PI and PC sequences on the grounds of groundwater composition. Along the line of traverse, profiles of As, Fe/Mn, V, and U (Fig. 7) delineate the regions of PI and PC aquifer.

Using colour to explore the subsurface lithology around drill sites and along the line of traverse between the drill sites, it was found that 94 % of black wells ($n=79$) confirmed the sedimentological setting for sites that had been deduced from drilling and water

chemistry, and 81 % of black wells contained $<10 \mu\text{g/L}$ of As (see table S1, [ESM](#)). The difference represents wells at the margins of palaeo-interfluvial settings where local migration of As-polluted waters has brought As to marginal PI settings and is now polluting wells that previously were low in As, as has been documented elsewhere (McArthur et al. 2010).

The combined colour and chemical data allowed mapping in detail of the contact between the regions of palaeo-interfluvial and palaeo-channel, which can be identified in the field to within a few metres by such means (e.g. Hoque et al. 2012). Along the central, PI, part of the traverse, local areas of red-stained wells in which well waters contains $>10 \mu\text{g/L}$ of As (sites 8, 13, 15, 17, 21), and low concentrations of U, V, and Mn. The depth of these wells is within the range 30–90 mbgl at which PI brown sands would be found if they were present. The fact that PC signatures are seen locally in isolated small areas suggests they represent local incision of minor palaeo-channels through the LGMP, i.e. local scour to the depth of the brown sand. Such regions have proven to be uncommon in the mapping.

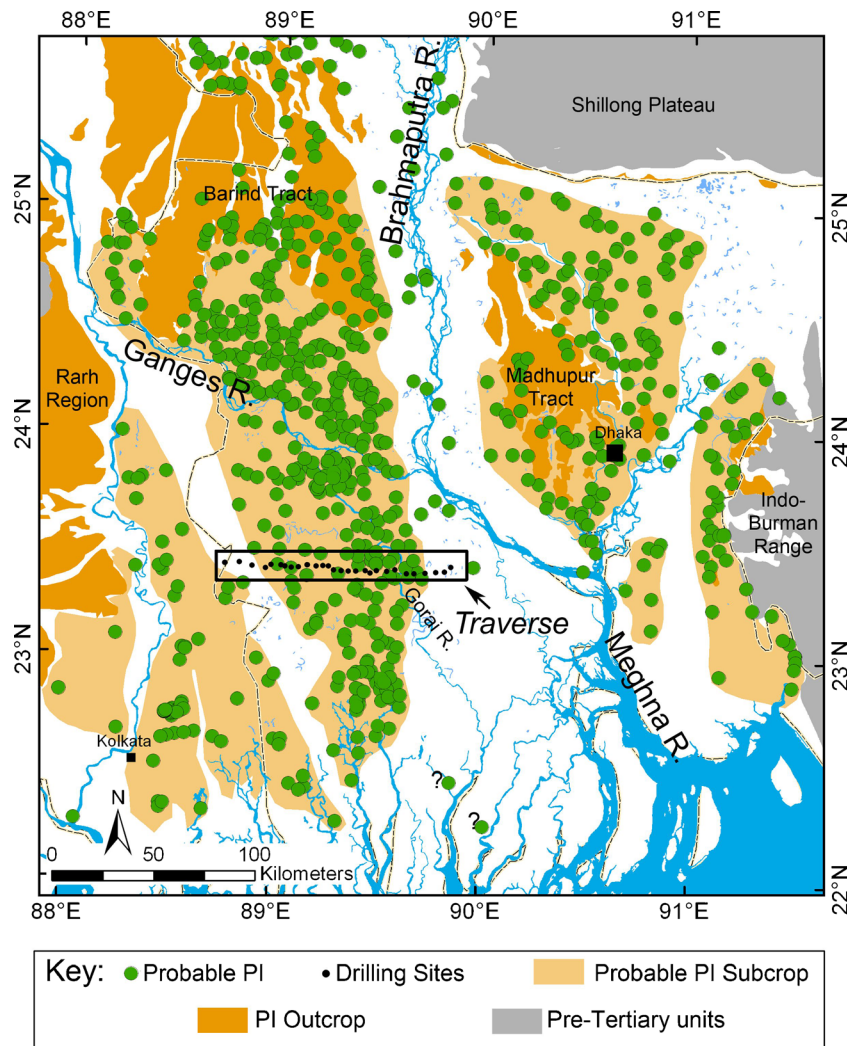


Fig. 4 Composite of Figs. 2 and 3, plus locations of brown sand reported in the literature. In this composite, sites of *Probable PI* based on water composition are lumped together with those based on historical logs and new drilling reported here. The figure shows in *light brown* colouration the predicted areas under which a PI sequence is probably present at depths between 20 and 90 m bgl in the Bengal Basin. The area covers around 45,000 km² of the Bengal Basin. Areas of *dark brown* as in Fig. 1. The putative PI sequences should host groundwater containing <5 µg/L As and typically much less. Note that palaeo-interfluves may be channelled locally at a scale too small to be visible here

Discussion

Extent of low-As PI aquifers

The analysis shows that low-As PI groundwaters that can be tapped from shallow depths accessible to local drilling methods are likely to be present under 45,000 km² of the Bengal Basin, which is one third of its area (Fig. 4). These resources of low-As groundwater have not been much exploited by agencies responsible for potable-water supply. They have been overlooked by many owners of private wells who typically drill wells to the shallowest depth at which water may be obtained. That depth is often less than the depth to the local palaeo-interfluvial aquifer.

Along the line of traverse, the sediments revealed by drilling (Fig. 5) indicates that the middle portion of the traverse does indeed comprise a palaeo-interfluvial sequence, as predicted from water composition and DPHE logs. At both traverse ends, this central PI sequence is bounded by grey sands of As-polluted PC aquifers which

occupy 40 km of the traverse. The post-LGM channels have not much truncated the PI sequence because their depth of incision has been mostly too shallow to do so, being imposed by decreased base-level gradients (Salter 1993) as sea level rose after 18 ka.

Fragmentation of low-As PI aquifers

The protection afforded by the LGMP, and the areal extent of PI aquifers, depends upon the degree of fragmentation of the late Pleistocene palaeo-interfluves. That fragmentation is governed by the initial distribution of rivers on that late Pleistocene landscape, and the degree to which subsequent channelling by post-LGM rivers has further fragmented those palaeo-interfluves. Such a large topic cannot be addressed fully here, but pointers exist that suggest such fragmentation is definitely not fractal in nature but instead has left many areas of PI aquifer that are tens of square kilometres in extent.

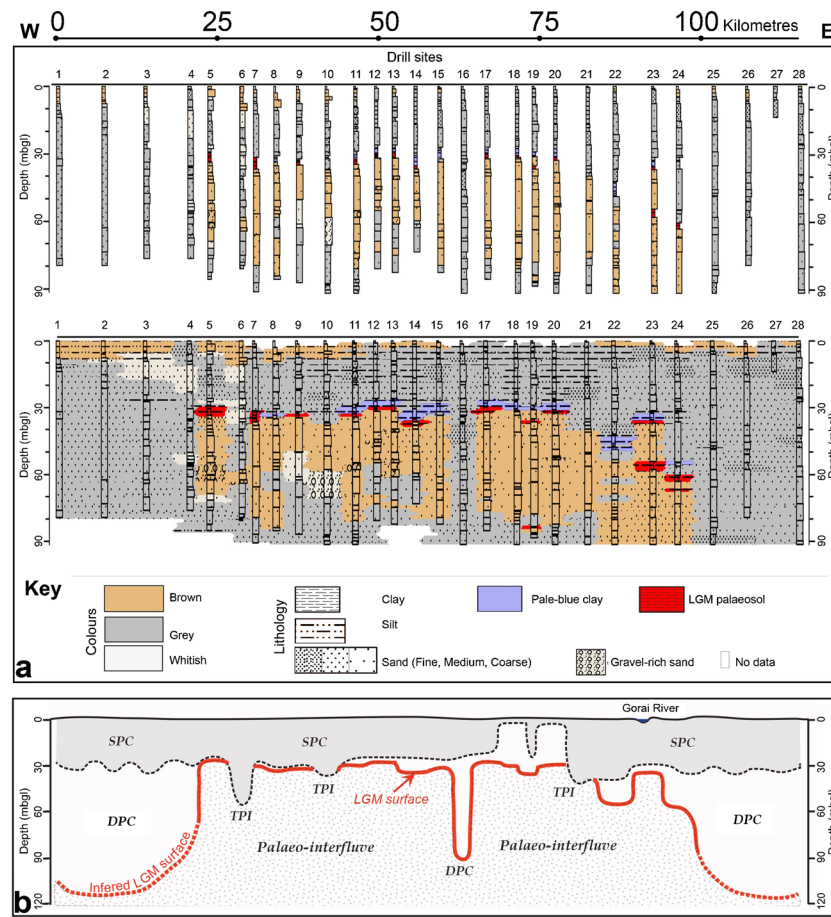


Fig. 5 Sedimentary sequences revealed by drilling along the traverse shown in Fig. 1. **a** Lithological records and geological section inferred from them. Note the variation in thickness of PI brown sand along the traverse. **b** Schematic topography of the land surface at the last glacial maximum (LGM) when sea level was some 120 m below that of today. *SPC* shallow palaeo-channel; *DPC* deep palaeo-channel, *TPI* truncated palaeo-interfluvial. Palaeo-channel and palaeo-interfluvial were defined by drilling; the location of boundaries between them was refined using hydrochemistry and survey in the field of the colour of stain on water wells (see text). Note the presence of two palaeosols at sites 19, 23 and 24. The upper is interpreted to be the LGMP

Whilst boreholes provide point-verification of subsurface lithology, the subsurface lithology between such tie-points can be reliably deduced from the composition of well water (McArthur et al. 2011; Hoque et al. 2012) and even from the colour of stain left by well water spilled on well completions (McArthur et al. 2011, 2012). Such investigations have shown that a borehole spacing of a few km (4–5 km in this study) reliably identifies major palaeo-channels. They have further shown that channeling at a finer scale has incised mostly to depths less than 30 m into the existing landscape, so preserving many of the buried PI sequences that mapped here. Where such incision has gone deeper, the fact is revealed by the presence of As-polluted wells that are screened at a depth greater than the depth to the local LGMP, which is typically at 25–30 m.

It can be observed that post-LGM channelling does not often incise to a depth greater than 30 mbgl, which suggests that much potential exists for PI sequences to have avoided erosion (Hoque et al. 2012). Nevertheless, sites 6 and 16 show that channelling at a scale of 1 km can incise through the LGMP and so fragment the areas of PI;

an important future task will be to delineate just how dissected the regions of buried PI aquifer are because the buried PI aquifers can be recharged only by lateral flow from adjacent palaeo-channels (McArthur et al. 2011, 2012). Data show that the spacing between such palaeo-channels is between 6 and 10 km.

It may be possible, in some areas, to delineate the PI and PC regions by examination of surface character using remote-sensing images as has been shown elsewhere (Hoque et al. 2012); for example, along the line of traverse, PI regions have a surface expression that lacks visible channelling (Fig. 8a), whilst PC areas show such channelling (Fig. 8b).

Protective nature of the LGMP

The value in the Bengal Basin of brown sand aquifers as low-As supplies of groundwater has been noted before (DPHE 1999; Pal et al. 2002; McArthur et al. 2004, 2008, 2011; Zheng et al. 2005; Stollenwerk et al. 2007; van Geen et al. 2007; von Brömssen et al. 2007; Pal and Mukherjee 2009; Burgess et al. 2010; Hoque et al. 2011, 2012). The local protection afforded to underlying aquifers in central Bangladesh by the residual

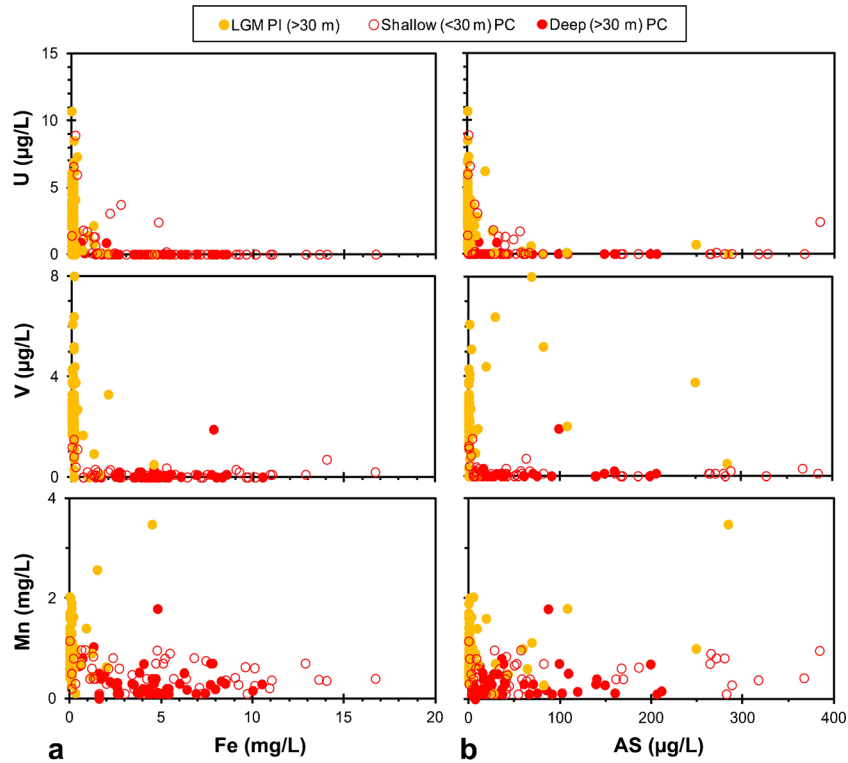


Fig. 6 Cross-plots of *As*, *Mn*, *Fe*, *V*, and *U* for groundwater from wells along the line of traverse shown in Fig. 1. The plots emphasize the mutual exclusivity of the element group *Fe* and *As* that characterise PC groundwaters, and the element group *Mn*, *V*, and *U* that characterize PI groundwaters

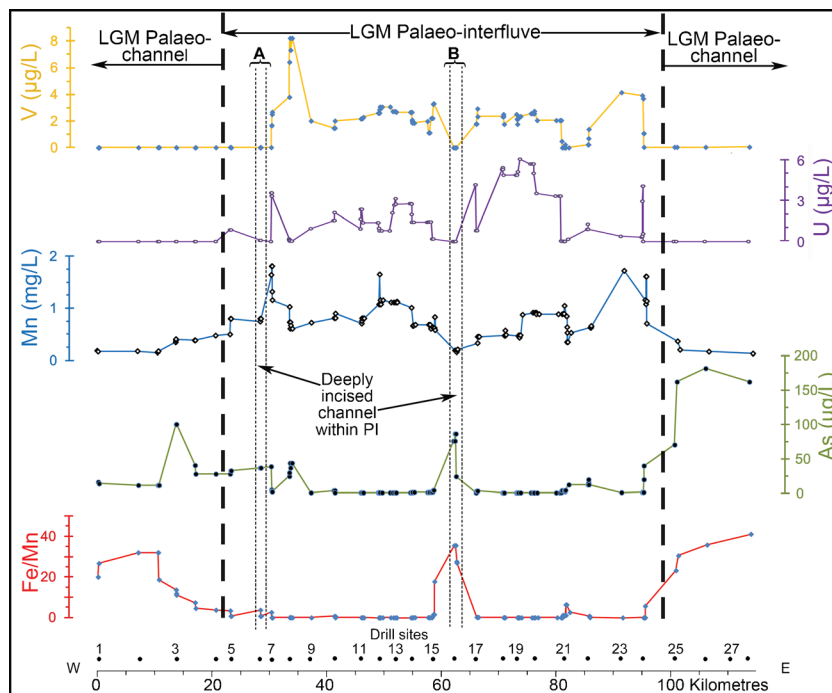


Fig. 7 Hydrochemical profiles along the traverse for wells between 20 and 91 m deep, shown as 5-point moving-medians. *At the bottom*, drill sites are *numbered* along the traverse. The PI areas are typified by low *Fe/Mn* ratios, low concentrations of *As* (mostly $\ll 10 \mu\text{g/L}$), and concentrations of *U* and *V* in the low $\mu\text{g/L}$ range. In PC regions, groundwaters contain $\text{As} > 10 \mu\text{g/L}$, little *Mn*, no *U* or *V*, and concentrations of $\text{Fe} > 0.9 \text{ mg/L}$. Using these criteria, a central region of palaeo-interfluve can be defined that is flanked at the traverse ends by major palaeo-channels, and is interrupted at *A* and *B* by minor palaeo-channels.

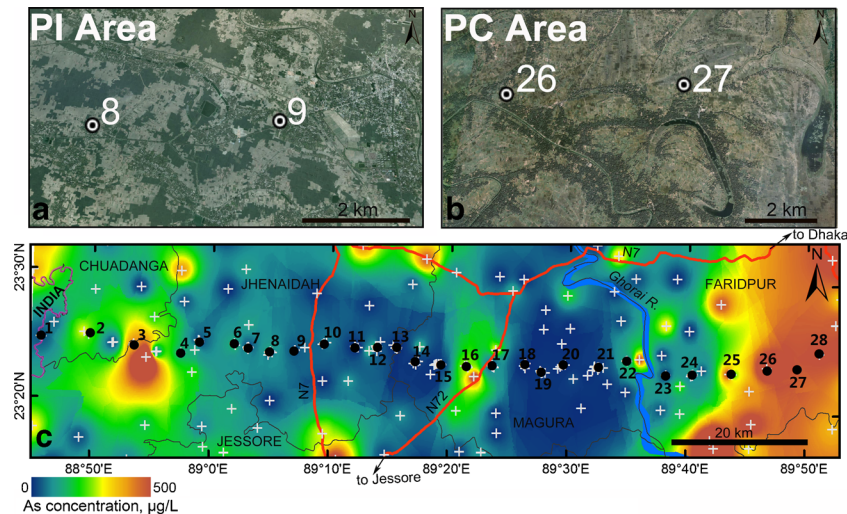


Fig. 8 a–b Satellite imagery (Google Earth Pro, with permission) showing surface features at a typical PI location (sites 8, 9) and at a typical PC location (sites 26, 27) on the traverse respectively. Regions of PI often lack surface channel-scars characteristic of PC regions. c Arsenic concentrations in groundwater along the traverse interpolated from the data (white crosses) of DPHE (1999, 2001) and this study (white crosses are covered by the black circles representing the drill sites). Drill sites numbered as 1–28. Roads are shown in red and boundaries of administrative units (districts) are shown in dark-grey

clays (palaeosols) of the Madhupur and Barind Tracts was recognised by Davies and Exley (1992) and Davies (1995). The more widespread role of the subcrop of the LGMP palaeosol in protecting aquifers across the Bengal Basin from As pollution was not, however, appreciated until relatively recently, following their documentation widely in the subsurface of West Bengal (McArthur et al. 2008, 2011; Hoque et al. 2012). The extent of the brown sand and capping palaeosol noted so far, however, represent but a minute fraction of the total area of the basin under which the data presented here suggests that such a sequence might lie.

This paper further confirms the palaeosol model of As pollution of groundwater (McArthur et al. 2008) which states that PI aquifers will be protected from downward migration of As in groundwater from overlying As-polluted aquifers by the impermeable LGMP. Thus, PI aquifers are likely to prove a long-term source of low-As groundwater.

Mitigation

The PI aquifers delineated in this study are suitable for mitigation of the As-pollution seen in PC aquifers. In many areas where the shallower SPC aquifers are currently exploited, they are underlain by PI aquifers, so mitigation would involve no more than drilling an existing well deeper so that its well-screen is emplaced in the brown sand of the underlying palaeo-interfluvium, and so beneath the protective LGMP. Indeed, this is already the default position in areas where the palaeo-interfluvial sequence is overlain not by sands, but by clays, silts, and peats, through which wells must be drilled to reach the first sands (e.g. sites 18 and 20; others in McArthur et al. 2011).

At the boundary of the PI and PC settings, the composition of well water can give confusing signals of sedimentology because of changing water composition caused by lateral migration of As-polluted groundwater from PC aquifers into

PI aquifers in response to natural flow and pumping, especially pumping for irrigation. Changes can be marked over a period as little as 2 years (McArthur et al. 2010). Low-As wells become As-polluted and black well-stain becomes overprinted by red stain, making colour differentiation difficult. Wells in marginal PI settings a few metres to a few tens of metres downflow of such confusing well waters frequently have extremely high concentrations of Mn (typically >2 mg/L), which imparts to the well and the well-completion an intense black stain that is unmistakable in the field (McArthur et al. 2011). This is the best marker of the proximity to the boundary between palaeo-channel and palaeo-interfluvium settings of wells, and allows boundaries to be mapped at the spatial scale of metres if wells are sufficiently common (Hoque et al. 2012; McArthur et al. 2012).

At PI margins, protection against lateral (horizontal) migration of As into PI aquifers is provided by sorption to sedimentary iron oxyhydroxide in the brown sands (Stollenwerk et al. 2007). Model estimates by that author for sorption coefficients (K_d) in Bangladesh sediments are around 30 L/kg. Measurements in Bangladesh at shallower levels (~ 6 m bgl), close to the zone of water-table fluctuation, indicate values of 7–8 L/kg (Jung et al. 2012). Push-pull, in situ, experiments in Bangladesh gave a value of 13 L/kg (Radloff et al. 2011). Assuming K_d value of 13 L/kg, a retardation factor 70 can be estimated for the brown sand of the Bengal Basin (taking 30 % porosity, 1.6 g/cm³ bulk density), and this value is in the range of field-based estimates of a factor of >30 by McArthur et al. (2010). Using this value in the Ogata-Banks solution (Ogata and Banks 1961), a well that is protected by a 50 m thickness of brown sand (e.g. one sited 50 m inside the margin of a PI sequence), would remain As free (<10 $\mu\text{g/L}$) for up to 525 years, assuming a groundwater velocity of 5 m/year, a hydrodynamic dispersion of $2e^{-7}$ m²/s, and an As concentration in

invading water of 100 µg/L. These estimates are tentative, given the paucity of in-situ measurements of retardation factors for As and the degree of latitude on all and any of the governing parameters. It nevertheless allows an estimate to be made of the frequency with which wells in PI settings need to be tested, unless at the PI margins, and suggests a decadal timescale would suffice.

The deep aquifer

An increased use of deep aquifer water (from depths >150 m) is often invoked as an alternative source of water that can be substituted for the As-polluted water of the shallow aquifers. A major concern about this proposal is the possibility that increased abstraction from the deep aquifers of the Bengal Basin may draw down As from overlying aquifers. Because the brown sand of the PI sequences has the capacity to sorb As, it is often taken to be a barrier to the downward migration of As into the underlying deep aquifer (Stollenwerk et al. 2007; Radloff et al. 2011). Such invocations seldom recognise the limited thickness of the brown sand, which rarely exceeds 30 m, and so far have never recognised that the LGMP will limit downward migration of water through brown sands and so reduce their effectiveness as a protective sorber.

Along the traverse, the pre-LGM brown sand is often thin (35 % is <20 m in thickness), and so has a limited capacity to delay downward migration of As, even were it to occur. Such downward flow from grey to underlying brown sand, however, cannot be widespread because brown sand is usually overlain by the LGMP and/or pale blue-grey clay (Fig. 5; McArthur et al. 2004; Hoque et al. 2012), the effective impermeability of which makes irrelevant, in most places, the sorptive capacity for As of the brown sand in respect of downward (vertical) migration of As. The sorptive capacity of brown sand may therefore be of little regional importance when assessing the vulnerability of the deeper aquifers to the downward breakthrough of As-rich groundwater from overlying aquifers in palaeo-channel settings.

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