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The palaeosol model of arsenic pollution of groundwater tested along a 32 km traverse across West Bengal, India

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ABSTRACT

The distribution of As-pollution in groundwater of the deltaic aquifers of south-eastern Asia may be controlled by the subsurface distribution of palaeo-channel sediments (As-polluted groundwaters) and palaeo-interfluvial sediments (As-free groundwaters). To test this idea, termed the palaeosol model of As-pollution, we drilled 10 sites, analysed groundwater from 249 shallow wells (screened <107 mbgl), field-tested another 149 for As, and used colour as a guide to the presence or absence of As-pollution in a further 531 wells. Our work was conducted along a 32-km traverse running W to E across southern West Bengal, India. At seven drill sites we logged a palaeo-interfluvial sequence, which occurs as three distinct units that together occupy 20 km of the traverse. These palaeo-interfluvial sequences yield As-free groundwaters from brown sands at depth<100 m. The palaeo-interfluvial sequences are separated by two deep palaeo-channels, which were logged at 3 sites. The palaeo-channel deposits host As-polluted groundwater in grey sands. Our findings confirm the predictions of the palaeosol model of As-pollution. We show again that well-colour can be used both to successfully predict the degree of As-pollution in groundwater, and to locate regions of buried palaeo-interfluve that will yield As-free groundwater for the foreseeable future.

1. Introduction

Much shallow groundwater in alluvial aquifers worldwide contains >10 μg/L of arsenic (As), the WHO guideline value for potable water (WHO, 2011): concentrations above 50 μg/L are common. The As threatens the health of millions of consumers; the problem is severe in the deltaic regions of South and East Asia (Berg et al., 2001; Fendorf et al., 2010; Ravenscroft et al., 2009; Winkel et al., 2008), where groundwater supplies most domestic potable water. The aquifers of the Bengal Basin are the most widely polluted, and pose the biggest threat to health (BBS/UNICEF, 2011) because of the large populations of Bangladesh (160 million) and West Bengal (60 million).

The As-pollution is a by-product of the reduction of sedimentary iron oxyhydroxides (Berg et al., 2007; Gulens et al., 1979; McArthur et al., 2001; Nickson et al., 1998, 2000; Polya et al., 2005 et seq.). As yet, however, only a partial understanding exists of the either the spatial distribution of this pollution process, or of the spatial distribution of the As it releases to groundwater; a better understanding would inform and aid development and remediation of the aquifers. We understand that As-pollution is minimal in areas of active recharge because rainwater contains little As, and because subsurface release of As by reduction of Fe-oxyhydroxides in areas of recharge is prevented by the dissolved oxygen in recharging water. Some areas of recharge have been identified conceptually (Khan and Hoque, 2002) or by geophysics (Aziz et al., 2008; Hoque et al., 2009; van Geen et al., 2006), or by drilling (Weinman et al., 2008) or by dating groundwater (Stute et al., 2007). Recharge is voluminous where the shallow aquifer is unconfined and crops out *e.g.* northern Nadia, West Bengal; for a map of such areas in Bangladesh, see Fig. 3 of Ravenscroft (2003).

In addition, we understand that groundwaters from aquifers comprised of brown sand are typically reported to be free of As pollution (Burgess et al., 2010; DPHE/BGS/MML, 1999; Hoque et al., 2011; McArthur et al., 2004, 2008, 2011; Pal et al., 2002; Stollenwerk et al., 2007; van Geen et al., 2003; von Brömssen et al., 2007), owing to the sorptive capacity for As of the Fe-oxyhydroxides they contain. Groundwaters from shallow grey sands are typically As-polluted because the original Fe-oxyhydroxide has been completely reduced and its sorbed As has been released to solution (DPHE/BGS/MML, 1999; McArthur et al., 2004, 2008, 2011; Stollenwerk et al., 2007; van Geen et al., 2003. The distribution of brown and grey sands, and so the distribution of As-pollution, may reflect the distribution of subsurface palaeo-interfluves and palaeo-channels (McArthur et al., 2008, 2011) that formed an ancient, now buried, landscape sculpted when sea-level was lower than it is today (Umitsu, 1987, 1993). We

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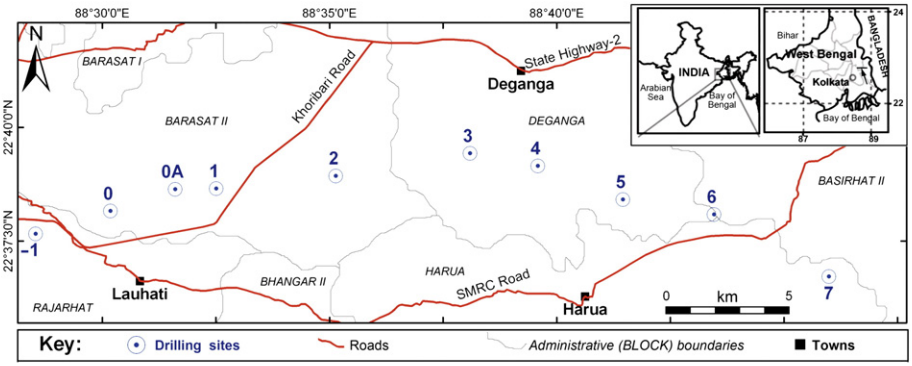

Fig. 1. Study area and drilling locations. Major roads and administrative boundaries (blocks) are included for reference.

report here a test of this idea through documentation of subsurface sedimentology, and its relation to As-pollution in shallow aquifers, along a 32 km traverse across south-central West Bengal. By shallow aquifers, we mean those tapped from depths of less than 150 mbgl, as defined by Ravenscroft (2003).

2. Evolution of the shallow aquifers, Bengal Basin

Sea-level decreased worldwide by ~130 m from 125 to 18 ka as glaciation developed to a maximum (Caputo, 2007; Rohling et al., 1998). The coastal areas exposed were channelled by rejuvenated rivers and palaeo-interfluvial (PI) areas were formed at an elevation around 30 to 50 m below the ground level of today (Goodbred and Kuehl, 2000; Umitsu, 1993). Increased hydraulic gradients promoted flushing of PIs by oxic water (Ravenscroft, 2003), whilst the exposed surfaces of the palaeo-interfluves were heavily weathered to form an extensive palaeosol of red clay. This clay is the laterite of Goodbred and Kuehl (2000), and the Last Glacial Maximum Palaeosol (LGMP) of McArthur et al. (2008). The rise in sea-level since 18 ka flooded the palaeo-valleys, which were infilled by grey sands that comprise some of today's aquifers in palaeo-channel (PC) settings. Later deposition of floodplain deposits buried both PI and PC sequences.

Groundwater from PI settings typically contains little Fe or As, and 0.2 to 0.4 mg/L of Mn, except around the PI margins, where Mn concentrations may reach 10 mg/L (McArthur et al., 2011, 2012). Groundwater from PC settings typically contains <0.2 mg/L of Mn, and is rich in Fe and As (McArthur et al., 2011, 2012). The LGMP prevents downward recharge of the aquifer in PI settings, which are recharged by lateral flow from palaeo-channels (McArthur et al., 2012). Palaeo-channel deposits, lacking the LGMP, are recharged by downward percolation through their confining beds, if any.

3. The study area

The study area comprises an E–W traverse across 32 km of southern West Bengal (Fig. 1). The traverse is located some 10 km to the

Fig. 2. Colour of stain on well-completions: (a) black, (b) black?, (c) red, (d) red?. Black coloration, from precipitation of manganese oxide, normally indicates both that a well water is As-free, and that it derives from a palaeo-interfluvial setting. Red coloration, from precipitation of iron oxide, normally indicates (in a well screened <150 mbgl) that the well water contains >10 μg/L As, and that it derives from a channel setting that may be predate or post-date the last glacial maximum (pre-LGM or post-LGM). Long well-screens, and a pronounced vertical stratification of groundwater composition, may mix Mn-rich and Fe-rich waters and so confuse this apparently simple division. Symbol (?) indicates ambiguous colour of a well-completion that is often clarified by viewing instead the colour of stain in a water-vessel *e.g.* the red-stained inside of the bucket in (d).

south of areas previously studied by us (McArthur et al., 2004, 2008, 2010, 2011, 2012), with a western end located around 3 km east of Kolkata Airport. Along the western part of the traverse, industrial-scale excavation since 1975 for brick-clay has removed up to 3 m of the upper aquitard (and agricultural land) and created depressions now used for aquiculture. These fish- and shrimp-farms are flooded by brackish water pumped from tidal creeks and rivers. Along the remainder of the traverse, scattered urban conurbations and villages are separated by agricultural land where irrigation with groundwater is ubiquitous. Across the study area, groundwater is the main source of water for drinking and domestic use, and for irrigation.

Beneath the present landforms, the palaeo-channel of the Ganges River follows essentially the present Hooghly River, some 10 km to the west of the western end of our traverse (Goodbred and Kuehl, 2000). The palaeo-channel of the present-day Ichamati River, in the eastern end of our traverse, also likely follows its present course.

4. Materials and method

We traversed E–W because major palaeo-channels and major palaeo-interfluves should be oriented N-S (Umitsu, 1993). We chose the line of traverse after linking the distribution of channel scars, visible across southern West Bengal on satellite imagery, to the distribution of As-pollution shown on maps posted on the website of the Public Health Engineering Department, Government of West Bengal (www.wbphed.gov.in, October 2010). Along the traverse (Fig. 1), we sampled 249 water wells that were screened at a depth < 107 mbgl. Samples were unfiltered except where visibly turbid. In the field, samples for laboratory analysis of cations and As were made 1% with respect to nitric acid. Laboratory analyses for Fe, Mn, and PO_4 were by ICP-AES; other analyses were done using a Bruker 90 ICP-MS. Using a Digital Arsenator®, we measured the concentration of As in the field in 149 well waters, of which 72 were analysed in the laboratory for other ions and As.

To supplement field-measurement of As, we recorded the colour of the well completions for 531 wells, classifying their colour as black, black?, red, or red?, where ? denotes ambiguity (Fig. 2). We did so because the colour of the completion, or domestic water-containers, can indicate both the degree of As-pollution of the source water and the presence of subsurface palaeo-interfluves: groundwater from PI settings contains dissolved Mn that stains well-completions black from precipitation of Mn-oxide; groundwater from PC settings contains dissolved Fe that stains well-completions red by precipitation of Fe-oxyhydroxides (see McArthur et al., 2011, 2012 for underlying rationale). The ambiguous colour of some well-completions may be clarified by viewing instead the colour of stain in a water-vessel *e.g.* the red-stained inside of the bucket in Fig. 2d.

Along the traverse we tested surface predictions of sedimentology by drilling 10 boreholes, about 3 km apart. Nine were drilled using the hand-operated reverse circulation method described by Ali (2003), and one deeper borehole (90 mbgl at Site −1) using commercial rotary drilling. Cuttings were logged and collected every 5 ft (1.52 m). The visualization package RockWorks® 14 was used to construct a lithological cross-section along the line of traverse. Using ERDAS Imagine® 8.4, we extracted a map of the surface expression of sedimentology (largely channel migration scars) from a Landsat TM image of September 2001. All locations are given to the WGS 84 co-ordinate system.

5. Results

5.1. Sedimentology

Lithological logs for each drill site, together with their interpretation, are shown in Fig. 3. Typical samples recovered by drilling are shown in Fig. 4. Further detail is given in the Supplementary Information. At three sites (0A, 4, and 5) we found a typical palaeo-channel sequence of grey sands, with overlying sequences comprising thin sands, silts, and clays. At Sites −1, 0, 1, 2, 3, 6, and 7, and at a site (not shown on figures) that was 200 m north-west of Site 0, we found a Late Pleistocene palaeo-interfluvial sequence, complete with capping Last Glacial Maximum Palaeosol (LGMP of McArthur et al.,

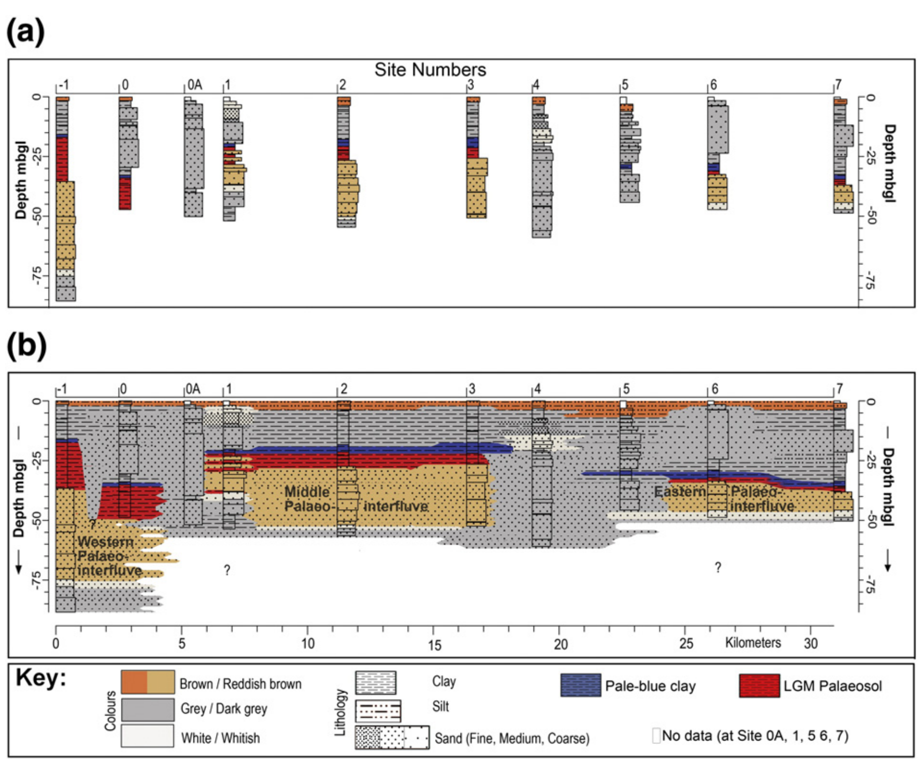

Fig. 3. Sedimentary sequences along the traverse: (a) lithological records along the traverse from west to east; (b) geological section inferred from (a) showing the palaeo-channel (PC) and palaeo-interfluvial (PI) zones, with zone boundaries defined by colour-mapping of wells and field testing for As in well water.

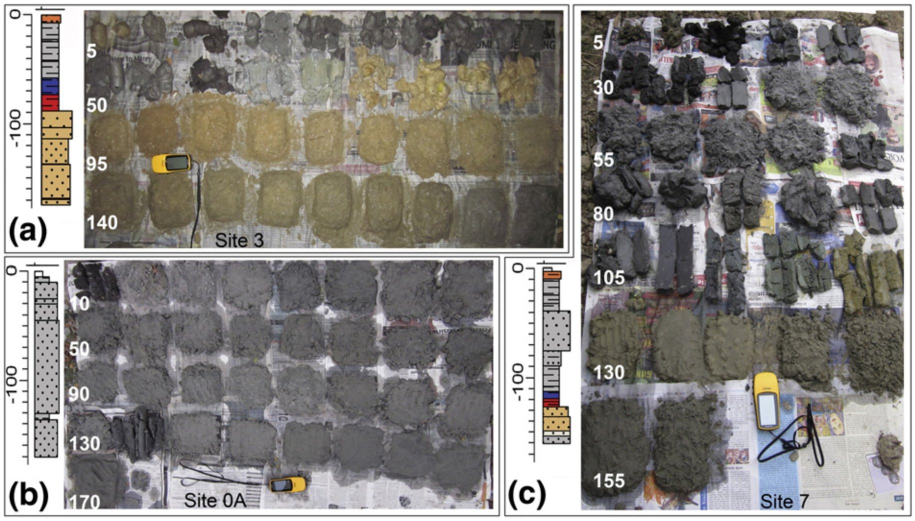

Fig. 4. Sediments recovered at drill sites locations in Fig. 1. Depth increases from top left to bottom right in increments of 5 ft. Lithological logs for all boreholes are given in Supplementary Information. (a) Site 3; a PI sequence where the last glacial maximum palaeosol (LGMP) at 80–95 ft is overlain by fine-grained silts and clays. The pre-LGM brown sands grade to grey at 180 ft bgl. (b) Site 0A; a deep PC record; grey sand throughout (record starts at 10 ft bgl); (c) Site 7; a PI sequence where the LGMP at 120 ft is overlain by post-LGM channel sands and silts.

2008). The LGMP sequence is everywhere overlain by a pale, blue-grey, clay, as found elsewhere (McArthur et al., 2004, 2008, 2011, 2012). At Site 5, this clay directly overlies, and is overlain by, grey sands. At Sites 0, 1, 6 and 7, the LGMP, and this overlying blue-grey clay, are in turn overlain by channel sands that form local aquifers.

The depth to the top of the LGMP differs between sites. Although apparently anomalously deep at Site 0 (35 mbgl), the depth-to-top gradually deepens eastward along the traverse. It is around 17 mbgl at Site −1 at the western end of the traverse and deepens to 36 mbgl towards its eastern end at Site 7. The thicknesses of the LGMP also differ between sites. It is thickest at the western end of the traverse (Fig. 3; Sites −1), where it reaches 20 m, and it thins eastward to 2.4 m at the eastern end of the traverse (Site 7). At all sites, the brown sands of the PI sequence grade downwards into grey sands. The thickness of the brown sand is around 38 m at Sites −1, 24 m at Sites 2 and 3 in mid-traverse, and 8 m at Site 7 at the eastern end of the traverse.

5.2. Water quality

The As concentrations in well-waters analysed in the field are given in Supplementary Information, along with the results for laboratory analyses of well waters, and the colour of well completions. For wells with black and black? completions, 68% have water with <10 µg/L As. For wells with red and red? completions, 57% have water containing >10 µg/L As. In Fig. 5, we plot the locations and colour of the 466 wells that are screened >30 mbgl (up to our deepest depth of 107 mbgl), and for which colour was recorded. We filter on depth in order to exclude the influence of around 60 wells that are screened in shallow palaeo-channels. The black and

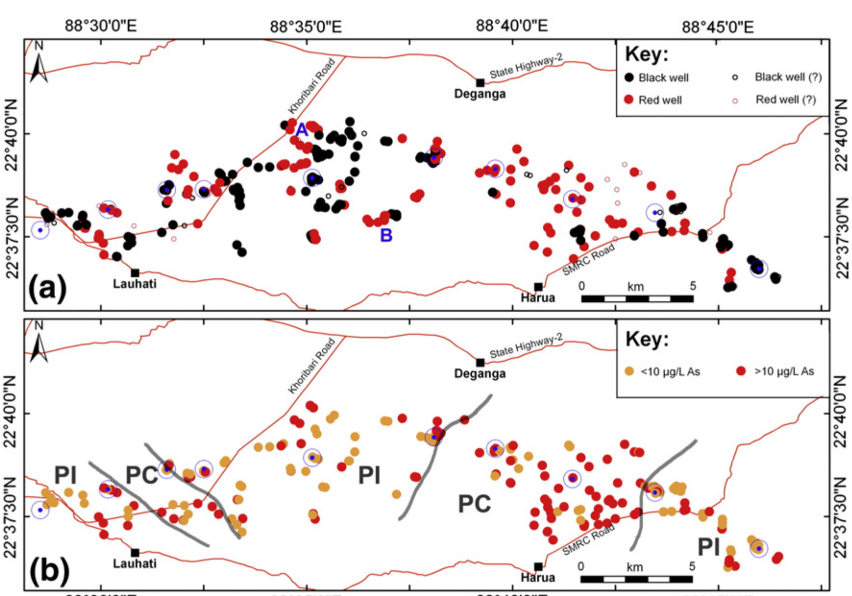

Fig. 5. (a) The colour of well completions along the traverse. To remove the influence of shallow, post-LGM channelling, we plot data only for the 485 of our 531 wells that are screened at >30 mbgl, and so likely below the base of any palaeo-channel sequence that may overlie any palaeo-interfluvial sequence. (b) Map of As concentrations for 248 wells in (a), and PI and PC settings are indicated.

black? completions cluster into three areas along the traverse (Fig. 5a), and red and red(?) completions mostly occur between them. Small clusters of red completions occur in areas where black is otherwise the predominant colour *e.g.* at A and B in Fig. 5a. The distribution of As (Fig. 5b), based on field and laboratory analyses for As in wells that are screened >30 mbgl, shows a similar pattern to that shown by completion-colour (Fig. 5a). Cross-plots of As, Mn, Fe, V, and U (Fig. 6) emphasise that PI groundwaters contain Mn, V, and U, whilst PC groundwaters contain Fe and As.

6. Discussion

6.1. The palaeosol model

The '*palaeosol model*', described elsewhere (McArthur et al., 2008, 2011), sets As-pollution into the context of sea-level change, weathering, and palaeosol formation. It is this model we test here. Along the 32 km traverse, groundwater composition, coupled to the sedimentology (Fig. 3) is consistent with the palaeosol model of As-pollution, which predicts groundwater rich in Mn, V, and U will be found in PI settings, and groundwater rich in Fe, As, and Mo will be found in PC settings (McArthur et al., 2012). This is what is seen in our data (Figs. 6 and 7). Our drilling (Fig. 3) and analysis identified three areas of PI sequence, separated by two deep palaeo-channels, one at Site 0A, the other at Sites 4 and 5. In all, some 20 km of our 32 km traverse is underlain by the full PI sequence of brown sands overlain by the LGMP. Groundwater hosted by these palaeo-interfluvial aquifers contain <5 μg/L of As (and mostly <2 μg/L). Conversely, where concentrations of As exceed 10 μg/L, concentrations of Mo and Fe are high, and those of U, V and Mn are usually low.

In mapping the extent of a PI sequence some 10–15 km to the north of our area, McArthur et al. (2011) showed that it was everywhere overlain by floodplain deposits — silts and clays, with intercalations of peat *sensu stricto*. At Sites −1, 2, and 3 along our traverse, the post-LGM sediments are similar fine-grained. At Sites 0, 1, 6, and 7, the post-LGM sediments are largely grey sands. These post-LGM sands typically yield groundwaters that exceed the WHO GV for As (10 μg/L) but not (in this study) the limit of 50 μg/L As set by the governments of Bangladesh and India for drinking water; the maximum we measured was 42 μg/L. Excepting around Site 7 (for reasons unknown), these post-LGMP grey sands are exploited for water supply in preference to the deeper, As-free, brown sands, the presence of which was unknown to inhabitants *e.g.* around Site 6. Mitigation of As-pollution in such areas would be accomplished by the simple expedient of drilling wells to be slightly deeper in order to exploit the As-free aquifer. If the situation described here is common across the Bengal Basin, that easy remedy could provide substantial benefit to many consumers in the short term.

Major palaeo-interfluves are separated by major palaeo-channels. The influence of minor tributaries in channelling and erosion has been largely overlooked. The existence of local erosion of the LGMP within the palaeo-interfluvial regions of our field area is suggested by the appearance of small clusters of red completions, containing >10 μg/L As, within regions of mostly black completions (*e.g.* A and B in Fig. 5). These high-As wells have screen depth (37 to 49 mbgl) that are below the local depth of the LGMP, and so are unlikely to be screened in brown sand. We suggest that these data reveal the presence of local, *i.e.* small scale, channelling by tributary rivers that locally cut through the LGMP and form small, local, palaeo-channel zones in which As-rich water occurs. A similar local channelling occurred at Chhota Jagulia, NE of Barasat, West Bengal (McArthur et al., 2011), where a small area of As-pollution occurs within a palaeo-interfluve, and in wells screened beneath the local depth of the LGMP.

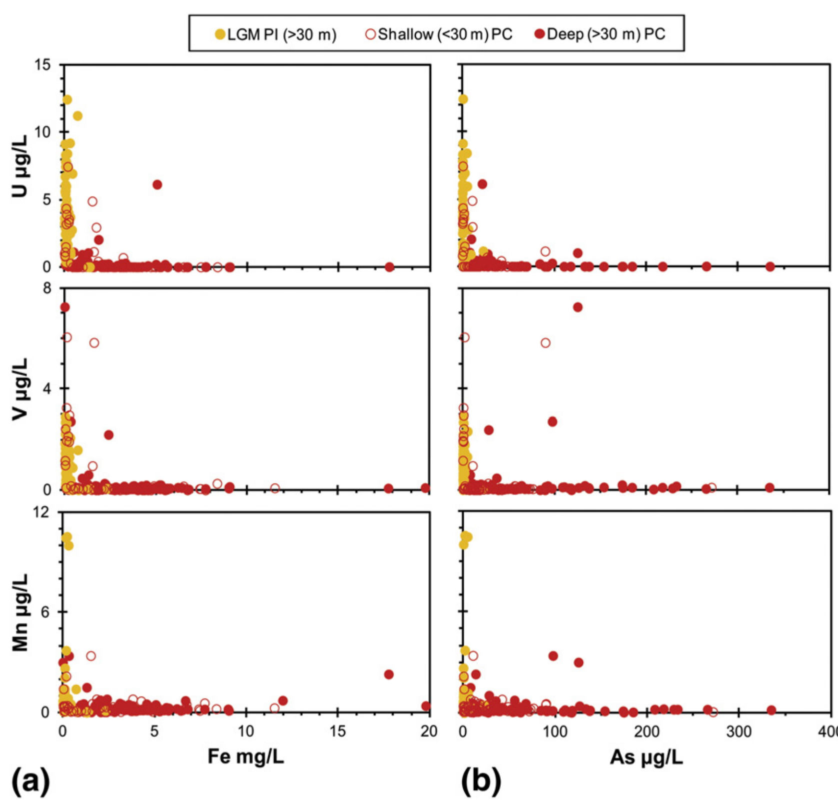

Fig. 6. Cross-plots of (a) Fe against Mn, V, and U; (b) As against Mn, V and U, showing the mutual exclusivity in solution of the group Fe, As, and Mo, which typifies PC settings of the aquifer, and the group Mn, V, and U, which typifies PI settings.

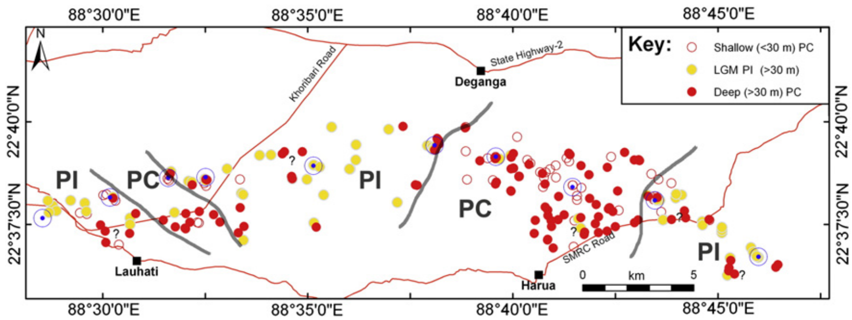

Fig. 7. Spatial pattern of palaeo-interfluvial (PI) and palaeo-channel (PC) water, differentiated on the basis of well colour survey, drilling results, and groundwater chemistry.

6.2. Landform and palaeosols

Extensive channel-migration features are seen on satellite imagery of West Bengal (Fig. 8). The lithological expressions are characteristic of the rapid lateral change in facies expected in a deltaic setting. The channelling revealed in Fig. 8 includes the two palaeo-channels that cut our PI sequences into three parts; channel-scars cross our traverse at Site 0A, and at Sites 4 and 5. The depth of incision of these palaeo-channels would have been dependent on relative base-level gradients (Salter, 1993). These decreased greatly as sea-level rose during the post-LGM period, especially from 18 ka to 6 ka. Our findings show that post-LGM erosion has not everywhere cut so deeply as to erase the LGMP, which we have found at depths from 17 to 36 mbgl, and has been found at similar depths elsewhere (McArthur et al., 2011). At Sites 0, 1, 6, and 7, post-LGM channelling did not cut-out the LGMP, but did scour deeply enough to form local channel aquifers of grey sand that overlie the LGMP. The timing of post-LGM channel formation cannot be deduced from this present work, although constraints have been placed on similar events elsewhere by C-dating (McArthur et al., 2004; Zheng et al., 2005; Sarkar et al. 2009).

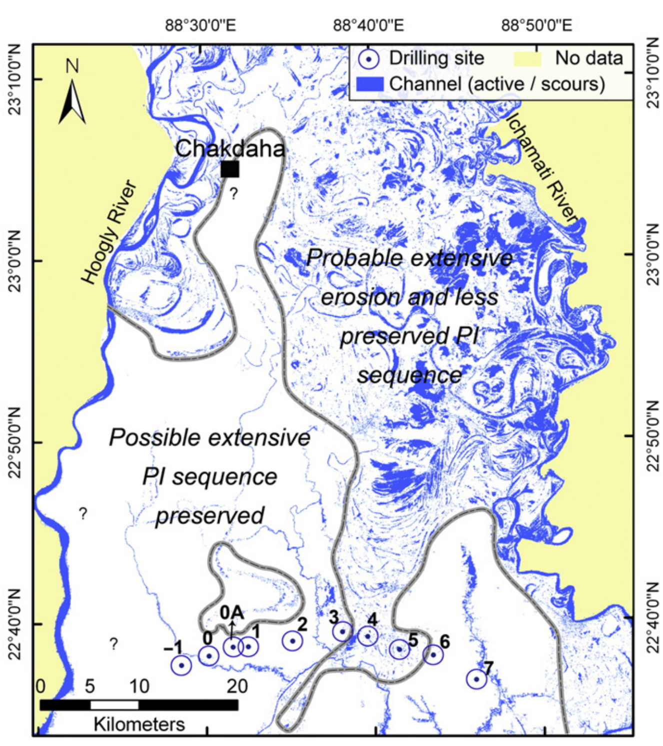

Fig. 8. Location of the modern-day channel and palaeo-channel scars derived from satellite TM image analysis, and their relation to our traverse. Symbol (?) indicates areas where image analysis is largely uncertain due to urbanisation. Chakdaha town was the study site for Biswas et al. (2012), which we discuss in Section 6.4.

The redness of the brown-sand sequence diminishes with depth and grades to grey at *ca.* 50 mbgl at all sites except Sites −1 (and possibly Site 0; see Fig. 3), where the transition is around 75 mbgl. The colour change indicates the base-level of the groundwater table at the time of active weathering up to the LGM at 18 ka. Below that base level, flushing by oxic water would have been slow, or the water would have been anoxic as it is today, and the sediments would not have been oxidised.

The brown sand is often presented as being a substantial barrier to arsenic migration because of its capacity to sorb As. Whilst that may be the case for *horizontal* migration of As in groundwater (McArthur et al., 2011) it is probably not the case for *vertical* migration, as is often invoked (Radloff et al., 2011; Stollenwerk et al., 2007). Along our traverse, the pre-LGM brown sand is thin (8 to 70 m) and laterally discontinuous. Regionally, this picture is probably repeated; the LGMP and underlying brown sand must have been punctured and/or thinned during the scouring of numerous palaeo-channels that are now filled by post-LGM grey sands. Thus, the thinness in places, and its absence in others, suggests to us that the role of the brown sand as a sorptive barrier to vertical migration of As into deep aquifers may have been over-emphasised. Furthermore, brown sand is, in many places, overlain by the LGMP, which is effectively impermeable (McArthur et al., 2008), a fact that decreases the importance of the brown sand when assessing the vulnerability of the deeper aquifers, which are composed of grey sands, to the breakthrough of arsenic-rich groundwater from overlying aquifers in palaeo-channel settings.

6.3. Palaeo-interfluvial margins

The contact between the regions of palaeo-interfluve and palaeo-channel can be identified in the field to within a few metres by colour-survey and field-testing of well-water for As (Fig. 9), as has been demonstrated elsewhere (McArthur et al., 2011, 2012). These contacts have been shown to be the locus of spatially-separated reduction fronts for Mn-reduction and Fe-reduction, where exceptionally high concentrations of Mn and Fe (and Mo and V) can be found (McArthur et al., 2012). The exceptionally high Mn concentrations at such fronts might prove a hazard to health (Khan et al., 2012; Wasserman et al., 2006), so it is important to identify such boundaries.

These PI/PC contacts are important also for the purposes of monitoring lateral migration of As-rich water into the palaeo-interfluvial settings of the aquifer from abutting palaeo-channel (McArthur et al., 2010, 2011). At the contacts, the As-polluted groundwaters flowing from the palaeo-channels lose As by sorption to PI brown sands, so it is across such boundaries that sentinel wells, regularly monitored, will allow quantification of the rate at which As in groundwater horizontally invades the presently As-free aquifers in their PI setting. At Site 3 (Fig. 9), in order to illustrate the ease and

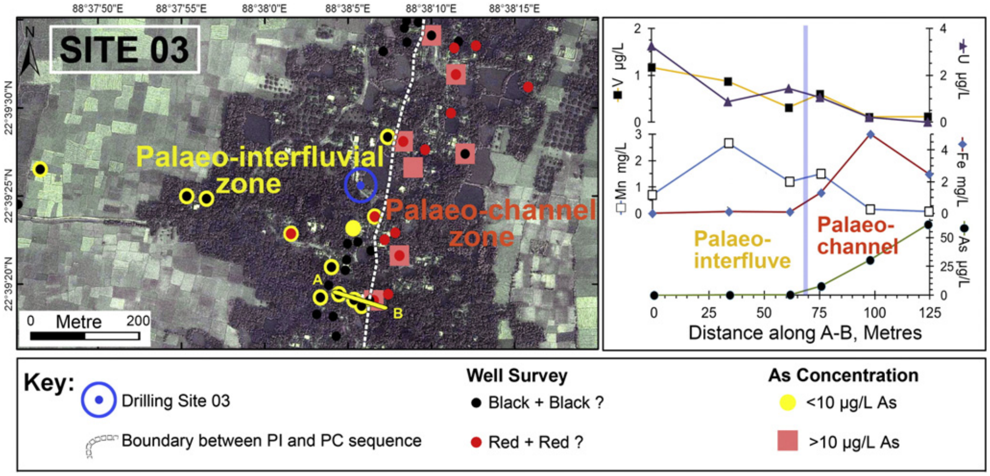

Fig. 9. Detail of boundary delineation between palaeo-channel and palaeo-interfluvial settings of the aquifer at Site 3. The boundary between PI and PC settings is arbitrarily set where concentrations of As go to zero.

precision with which the PI/PC contact can be established, we quantified its position to within 10 m along a line of traverse.

6.4. Other areas

Along the line of traverse reported on here, the PI sequence is overlain at Sites −1, 2, and 3, by a low-energy sequence of silts and clays, as has been found further north between Barasat and Habra (McArthur et al., 2011). Elsewhere along the traverse, the PI sequence is overlain by shallow palaeo-channels that had scoured deep enough to provide an exploitable aquifer, but not deep enough to erode the LGMP. In such a setting, an upper aquifer of grey sands containing As-polluted groundwater is separated by the LGMP from a slightly deeper aquifer of brown sands (within 100 mbgl) that host As-free groundwater. This shallow, dual-aquifer, system occurs elsewhere, but in a context where the brown sands have been termed the 'deep' aquifer (Van Geen et al., 2007; Zheng et al., 2005). We restrict the term 'deep aquifer' to mean those hosting groundwater below 150 mbgl (DPHE/BGS/MML, 1999; Ravenscroft, 2003).

The shallow, dual-aquifer system may be more common than appreciated. One area in which it might occur is Nadia District, West Bengal. The colour-screening method of McArthur et al. (2011) for

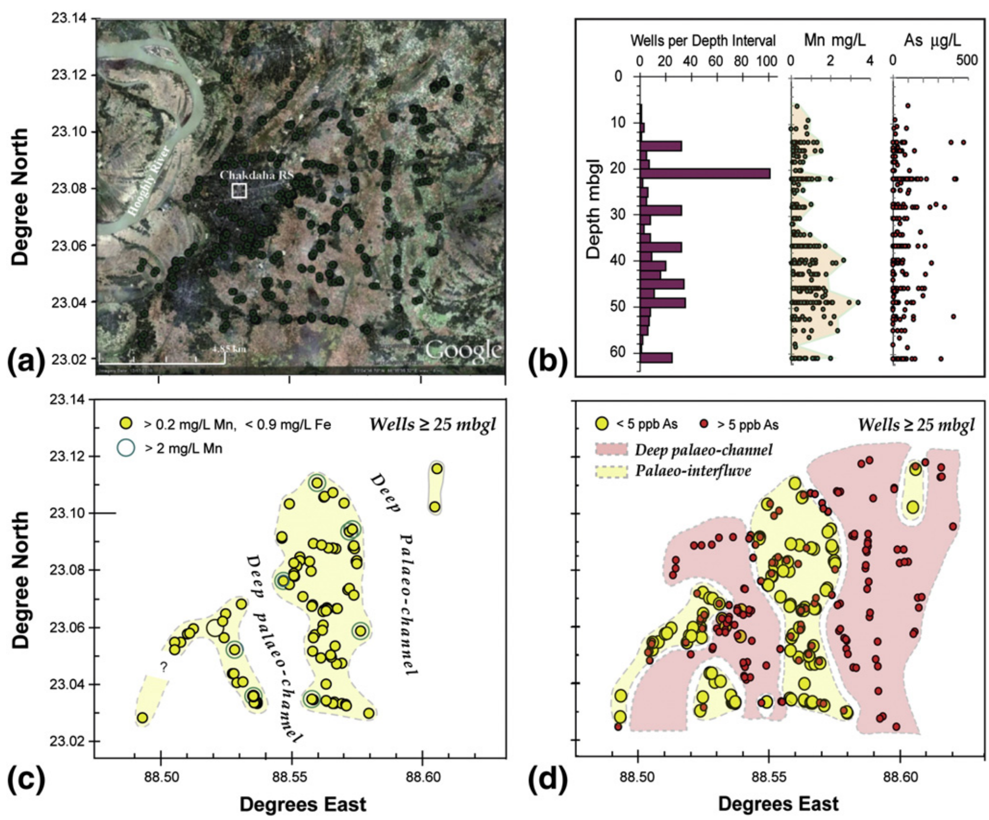

Fig. 10. Analysis of the data of Biswas et al. (2012). See text for interpretation. (a) Field area of Biswas et al. (2012). (b) Histogram of well depths, showing that a maximum number of wells are screened at 20–22 mbgl, and that a minimum number are screened at 22–24 mbgl, plus depth profiles of As and Mn. For Mn, concentrations reach higher levels below 30 mbgl than above it. (c) Distribution of well waters at depth ≥25 mbgl with >0.2 mg/L Mn and <0.9 mg/L Fe, together with locations of well-waters containing >2 mg/L Mn. (d) Distribution of well waters with depths ≥25 mbgl and with <5, or >5, µg/L of As.

mapping As-pollution in groundwater has been used by Biswas et al. (2012) to map As-pollution around Chakdaha, Nadia District, West Bengal. In presenting the method, McArthur et al. (2011) made clear that its secondary purpose was to map As-pollution, and that its primary purpose was to reveal the subsurface distribution of palaeo-interfluvial and palaeo-channel sequences. Biswas et al. (2012) did not use it for that primary purpose so, with their data, we do so here.

Of the 423 wells reported by Biswas et al. (2012), no fewer than 101 (24%) are between 20 and 22 m in depth (Fig. 10b). Only two (0.5%) are 22–24 m deep. We speculate that the level around 22–24 mbgl may be known to the area's drillers as a local base to the shallow aquifer above which drilling should stop. We further speculate that this may be because that is the depth to the LGMP in some areas in which it may be present. In contrast, maximum concentrations of Mn are higher below 30 mbgl than above that level, possibly indicating that a PI sequence may lie deeper than 30 mbgl, rather than below 22 mbgl (Fig. 10b).

As groundwater from palaeo-interfluvial settings tends to contain >0.2 mg/L of Mn and little Fe (here we take <0.9 mg/L; McArthur et al., 2012), we show in Fig. 10c the geographical distribution of wells with depths >25 m that contain >0.2 mg/L Mn and <0.9 mg/L Fe. The wells cluster into three main areas, which we postulate are subsurface PI sequences (a similar result is obtained for wells screened at>30 mbgl). Confirmation that a PI sequence occurs at depth comes from the distribution of As in water from wells screened at depths >25 mbgl (Fig. 10d). Well waters with <5 μg/L As, and so likely to derive from brown sands of a PI sequence, show a similar clustering to that shown in Fig. 10c for low-Fe, high-Mn waters. The putative PI regions do have within them some wells that are >25 m deep, and contain higher amounts of As, so it is possible that the PI sequences we identify are truncated *i.e.* regions where the LGMP itself, but not much of the underlying brown sands, have been removed by post-LGM channelling. In contrast, concentrations of Mn> 2 mg/L appear to flank the margins of the putative PI sequence, as would be expected if full PI sequences are present because the PI/PC contact at the margin of PI sequences is often the locus of Mn- and Fe-reduction fronts that drive concentrations to extreme values (McArthur et al., 2012). We know of no drill-sites in the areas of the postulated PI sequences with which to test these inferences.

The distribution of data for well waters around Chakdaha is that expected from a palaeo-interfluvial sequence that was deeply dissected by major palaeo-channels, and later scoured and reworked by Recent river channelling. We interpret the areas between the putative PI settings as being deep palaeo-channels (up to 61 mbgl, the maximum well-depth recorded), on the evidence that high As concentrations are to be seen at all depths in the area (Fig. 10b). Finally, high concentrations of As are found in shallow wells (<25 mbgl) across the entire area, showing that the subsurface PI sequences are covered by As-polluted aquifers comprised of shallow palaeo-channels.

7. Conclusions

The data we present fail to falsify the 'palaeosol model' for the distribution of As-pollution in the Bengal Basin. Similar testing should now be implemented in other As-affected areas of the Bengal Basin, and other deltaic aquifers, in order to establish how widely the model may apply. Wider testing will be assisted by adopting the colour-survey method of McArthur et al. (2011) that can, literally at a glance, distinguish between As-free wells that tap palaeo-interfluvial sands and As-polluted wells that tap palaeo-channel sands. Analysis of groundwater for V, Mo, and U, as well as the commonly measured Fe, Mn, and As, can be used to confirm the results of a colour survey.

How much PI sequence should we expect to find across the Bengal Basin? Erosion and sea-level change have operated on a 100 ka timescale since 800 ka (Lisiecki and Raymo, 2005): the cycle we examine is but the latest of many. It is therefore no surprise that Hoque et al. (2011) recorded a patchy occurrence of brown sands (relict PI) at depth in the central Bengal Basin. With respect to the latest cycle (125 ka to now), the amount of PI preserved will depend upon the depth and extent of post-LGM erosion. That, in turn, depends upon the base level of rivers (which is governed by sea level), the pattern of erosion since 18 ka, and basinal rates of subsidence.

Rates of subsidence are highest in the south-east of the basin, and in the Sylhet sub-basin of north-eastern Bangladesh. They are lowest at the basin margins (Khan and Hoque, 2002; Steckler et al., 2007), the western margin of which is close to the west end of our study area. Our finding of subsurface PI sequences in the western part of the basin, where relative subsidence rates are low and erosion potential therefore high, implies that much subsurface PI may be preserved by subsidence in the more rapidly subsiding parts of the basin. The finding of brown sand at depths of 195 to 229 mbgl at Sreenagar, central Bangladesh (Hug et al., 2011), close to the banks of the present Ganga River, the possible identification of PI at Chakdaha, and the finds reported here of LGMP at 36 mbgl, also suggest that subsurface PI sequences may be more extensive than commonly supposed.

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Appendix A. Supplementary data

Lithological logs, well-survey data, results of field-testing for As, and laboratory analysis of groundwater, are given in Supplementary Information. Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.05.038.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.05.038.

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