Arsenic in groundwater: the deep Late Pleistocene aquifers of the western Bengal Basin.

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ABSTRACT: in groundwaters from 145 wells across central West Bengal, India, those from Pleistocene aquifers at depths > 70 m beneath palaeo-interfluves contain < 10 μ g/L As. Pleistocene aquifers beneath deep palaeo-channels typically host groundwaters containing 10 to 100 μ g/L As at depths between 120 and 180 m. The depth profiles of As and SO₄, and the conservative tracers Cl/Br, δ^{18} O, and δ^{2} H, show that the As in Pleistocene groundwater beneath deep palaeo-channels is relict and does not arise from migration downwards of As-polluted groundwater in overlying aquifers. We postulate that the As was liberated *in-situ* by reduction of minimal iron oxyhydroxides in the grey Pleistocene sands by organic matter infiltrating from river-beds during late Pleistocene or earliest Holocene times. Mitigation of the widespread As-pollution in shallow aquifers through exploitation of deep Pleistocene aquifers would improve if guided by an understanding of the distribution of buried palaeo-channels and palaeo-interfluves and the knowledge that As may be present naturally in groundwater at depths > 150 m beneath deep palaeo-channels.

Keywords: arsenic, pollution, groundwater, West Bengal, aquifers, deep.

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1. DEEP and SHALLOW ARSENIC

Across Bangladesh and West Bengal, water-wells tapping shallow aquifers, typically at depths less than 60 m, provide the domestic water supply for much of the combined populations of 250 million. At such depths, concentrations of arsenic (As) frequently exceed the World Health Organization's Guideline Value of $10 \mu g/L$ for drinking water.^{1–4} Such water is termed here As-polluted. As late as 2009, some 52 million people in Bangladesh⁵ and 26 million people in West Bengal⁶ consumed such groundwater, which adversely affects health and may prove fatal.^{7–13}

The As-pollution (As > 10 μ g/L) is largely confined to groundwater in aquifers of grey sand that were deposited onto a late Pleistocene landscape as sea-level rose after the last glacial maximum (LGM) at 18 ka; for convenience here, these aquifers are termed 'Holocene' despite the fact that their basal sands might be as old as 18 ka (Section S1of the Supporting Information, hereinafter SI). Groundwaters in Pleistocene aquifers underlying the late Pleistocene land-surface usually contains < 10 μ g/L As^{3,4,14-20} and, as a consequence, wells in Pleistocene aquifers increasingly replace shallow wells in Holocene aquifers as a water source in order to avoid shallow As-pollution. The depth to the Pleistocene aquifer varies regionally within the basin and, although poorly known, is widely held to be no more than 150 m (Ref. 3 *et seq*). The default option is therefore to install a deep well, *i.e.* at a depth > 150 m, in order to ensure it is located in a Pleistocene aquifer hosting groundwater containing < 10 μ g/L As. In Bangladesh, deep wells have become the *de facto* preferred option for mitigation despite other options being favoured by government policy. ^{13,21,22} In West Bengal, community wells 152 m deep (500 ft) were adopted in the late 1990's by the state government as a mitigation option.

Across the Bengal Basin, a small but significant number of deep wells (*i.e.* > 150 m depth) have been reported to contain more than 10 μ g/L of As^{3,4,22–26} (see also www.wbphed.gov.in, accessed 28 08 2014). These occurrences raise concern that As is migrating to depth in response to deep abstraction of groundwater for irrigation and domestic supply.^{3,4,17,26–28}. In West Bengal the concern is acute, given the use there by government agencies since the 1960s of abstraction of deep groundwater for irrigation on an industrial scale (Ref. 29 and TOC Art). In the long-term, downward migration of As from the shallow aquifers may compromise the value of the deep aquifer as a source of low-As water. Here, using the spatial distribution of As, and the conservative tracers Cl/Br, δ^{18} O, and δ^{2} H, we attempt to constrain the effect on deep groundwater in Pleistocene aquifers of downward migration of shallow As in Holocene aquifers and explain the relevance of our finding to As-mitigation.

2. FIELD AREA

The field area comprises 235 km² of the western Bengal Basin between latitudes 22.9° and 23.1° and longitudes 88.5° E to 88.7° E (Fig. 1a) in West Bengal. Previous work in this same study area³⁰, based largely on drilling, defined the extent and depth of palaeo-interfluves and palaeo-channels and showed how they controlled the distribution of As-pollution of groundwater at depths < 70 mbgl. In this paper, we deal with the distribution of As-pollution in late Pleistocene aquifers > 70 m deep, all of which are composed of grey (not brown) sand, and all of which underlie either palaeo-channels or palaeo-interfluves. For details of the distribution of palaeo-channels and palaeo-interfluves, see Ref. 30, references therein, and Section S2 of the Supporting Information (SI) where an East-West cross-section through the area, Fig. S1, is given along the profile line A–A on Fig. 1a.

The study area comprises a buried late-Pleistocene landscape of palaeo-interfluves and palaeo-channels that is overlain with mostly grey sands deposited since the termination of the last glacial maximum (LGM) at 18 ka. The top of the palaeo-interfluves is typically between 35 and 38 metres below ground level (mbgl), based on drilling of 73 boreholes³⁰. The depth to the bases of the palaeo-channels is ≤ 70 mbgl, a figure based both on the depth to the base of the oxidised brown sands underlying the palaeo-interfluves^{20,30} and considerations of land elevation and sea-level lowering up to the time of the LGM (see Section 1 of the SI for an extended discussion of these points). We acknowledge that neither OSL nor ¹⁴C dates are available from the study area to validate the depth to the LGM surface.

Palaeo-interfluves in the Bengal Basin are, by definition, capped with a palaeosol, termed here the last-glacial maximum palaeosol, or LGMP.³¹ The presence of the LGMP defines the extent of two palaeo-interfluves that cross the study area from north to south (Fig. 1a). The LGMP forms an effectively impermeable barrier to downward flow of water³¹ so palaeo-interfluvial aquifers are recharged at palaeo-interfluvial margins by focussed flow of groundwater from adjacent palaeo-channels^{31–33} (see also Fig. S2). The LGMP is underlain by Pleistocene sand, the uppermost 30 m of which is brown through oxidation during LGM times^{3,20,30} Deeper grey sands were not oxidised during LGM times. Groundwater from the grey sands only was sampled here.

Palaeo-channels in the Bengal Basin have a range of forms simplified here for clarity into two main types. The main river-channels of LGM times now infilled with grey, mostly Holocene, sands are termed here deep palaeo-channel aquifers; their groundwaters are widely polluted by As. These aquifers are at depths between 35 and 70 mbgl and termed deep to distinguish them from shallow palaeo-channel aquifers which are at depths between 0 and 35 mbgl. The latter were formed by reworking during river avulsion of the uppermost 35 m of sediment since 6 ka, which has left a near-continuous blanket of post-6 ka shallow paleo-channel sands covering both palaeo-interfluves and deep palaeo-channels across most of the study area and much of the southern and central Bengal Basin. ^{19,20,30}

3. SAMPLING and ANALYSIS

During the period February 2011 to February 2014, we sampled 145 wells > 70 m depth tapping groundwater in Pleistocene aquifers of grey sand sited beneath both deep palaeo-channels and beneath palaeo-interfluves (Fig. 1; Table S1 and Figs S1, S2, of the SI): the locations of wells are referenced to the WGS 84 co-ordinate system. Many were private hand-pumped wells. Most others were hand-pumped community wells fitted with Mark II piston-pumps, installed by the government of West Bengal between 1986 and 2013 in order to mitigate As-pollution, and at a intended depth of 152 m (500 ft) depth with screen lengths of 11 m (36 ft). For 24 deep wells, the pump was removed and depth measured including the 8 wells, all Mark IIs, which had elevated Cl/Br in their waters. These 8 wells were sampled through a plastic pipe, 5%- inch in diameter and 150 foot long, that was inserted down the well after removal of the pump assembly. We also sampled two well-nests installed in 2011 in Dasdia at sites MP and SP (Fig. 1a). Site SP is in a palaeo-channel < 10 m from the edge of a palaeo-interfluvial margin. Site MP is 120 m into the palaeo-interfluve west of SP and so near a palaeo-channel margin. We analyzed groundwaters for As, Cl, Br, SO4, δ18O and δ2H by methods given in Section S3 of the SI.

4. RESULTS and DISCUSSION

Spatial Distribution of As. In our study area, groundwaters from beneath palaeointerfluvial interiors contain $\leq 8~\mu g/L$ of As but shows a range of concentrations, with the highest clustering around depths of 50 m and 150 mbgl (Figs. 2, S3). In contrast, only 8 groundwaters from beneath deep palaeo-channels contain $< 10~\mu g/L$ As (Fig. 1b; Table S1); the rest contain concentrations from 10 and 100 $\mu g/L$ As, with most high concentrations clustering around depths of 30 mbgl and 130 mbgl (Figs. 2, S3).

Establishing the reality of the maximum As concentration at 130 m is compromised by the lack of wells in the depth range 65 to 90 mbgl. Nevertheless, the As profiles in our well-nests at SP and MP accord with this deep maximum, and the minimum around 90 m depth (Section S4 of the SI). Furthermore, concentrations of As in groundwaters across Bangladesh^{3,4,34} and West Bengal^{35,36} are highest between 20 and 40 mbgl and decline with depth thereafter to depths around 90 m. These similar concentration profiles exists despite deep pumping for irrigation being common in West Bengal since the 1960s but uncommon in Bangladesh. The profiles suggests that drawdown of As may be less than commonly presumed and that the low concentrations of As around 65 – 90 mbgl are real.

The depth profiles show that As concentrations reach 150 μ g/L in some groundwaters from beneath palaeo-interfluvial margins (Figs. 2, S3). This is not unexpected, given that aquifers beneath palaeo-interfluves are partly recharged from As-polluted palaeo-channels, but the fact that As-pollution of groundwater is not found everywhere under palaeo-interfluvial margins (Fig. 1b) shows that regional flow into the palaeo-interfluves may be enhanced by local pumping *e.g.* where a high-yield irrigation well is sited inside a palaeo-interfluve and close to its margin.

Groundwaters in Pleistocene aquifers beneath palaeo-interfluves are essentially free of As, whilst those in Pleistocene aquifers beneath deep palaeo-channels are mostly As-polluted. This deep As may have three origins. Firstly, it may have been drawn down from As-polluted *shallow* palaeo-channel aquifers by deep pumping, although the minimum at ≈ 90 mbgl in the depth profiles of As and SO₄ (Figs. 2, S3) make this unlikely. Secondly, it may have been drawn down from overlying As-polluted *deep* palaeo-channel aquifers by deep pumping. Thirdly, it may have been generated *in-situ* at or close to its present location and depth. We examine these three possibilities below.

Cl/Br and SO4. A plot of Cl concentrations versus Cl/Br mass ratios for groundwaters allows discrimination of sources of Cl. The plot can distinguish waste-water contributions to groundwater from those made by brackish connate-water of marine origin.^{37–39} Using Cl/Br, shallow palaeo-channel aquifers in the study area⁴⁰, and elsewhere in West Bengal³³, have been shown to be pervasively contaminated by Cl from waste-water. They are also pervasively polluted by As.³⁰ Mixing with brackish connate water elevates concentrations of Cl above 10 mg/L in a few deep groundwaters, but these are identifiable because they plot on the brackish mixing line (Fig. 3).

From the above, it is clear that shallow groundwater and its As-pollution could not invade deep Pleistocene aquifers of the study area without being revealed by the presence of more mobile contaminant Cl and an elevated Cl/Br. Yet none of our groundwaters from Pleistocene aquifers beneath palaeo-interfluvial interiors have elevated Cl/Br (Fig. 3) and so none contain shallow groundwater or its As-pollution. This observation is consistent with the conclusions of the previous section.

Around 72 % of groundwaters from beneath deep palaeo-channels have Cl/Br ratios that plot on or below the mixing line between recharge and brackish connate water (Fig. 3), suggesting that they are uncontaminated by Cl or As in shallow groundwater. These 72% of groundwaters nevertheless contain typically 10 to 100 μ g/L As with outliers up to 326 μ g/L (Table S1). Some 28% of groundwaters from beneath deep palaeo-channels have elevated Cl/Br. A Cl-mixing model (Section 6 of the SI) assigns a contribution to these groundwaters from As in shallow groundwater of \leq 16 μ g/L As and mostly very much less. Thus, little of the As, and that in only 28% of our deep groundwaters, is likely to be derived from shallow groundwater.

Furthermore, groundwaters show an inverse relation between Cl/Br and As (Fig. S4). They do so because waste-water has elevated Cl/Br and contains many mg/L of Cl, NO₃, and SO₄ ⁴⁰. The NO₃ suppresses reductive dissolution of iron oxyhydroxides and As-release. The SO₄ sequesters As from solution into neoformed pyrite. Our analysis may therefore overestimate the contribution to deep As that could be made by shallow groundwater.

Some deep groundwaters with < 12 mg/L Cl plot below the brackish-water mixing-line (Fig. 3). They do so for one or more of three reasons: they have received additional Br from organic decay; 33,40,42 they have suffered evaporative concentration during recharge; the end-member, chosen to represent the freshest shallow groundwater, is inappropriate for deep groundwaters. Addition of organic Br can displace Cl/Br downwards from positions above the mixing line or from positions on the mixing line. If the former, the Cl mixing-model shows that they contain no more than 4% of waste-water, with amounts up to 9% for the two highest Cl concentrations. These additions would carry < 6 μ g/L of As (Section 6 of the SI) and so contribute little to As-pollution.

Further constraint on downward movement of groundwater can be obtained from a consideration of the depth profile of SO_4 . Below 70 mbgl, most concentrations of SO_4 are below our detection of 0.01 mg/L. Where SO_4 is found below 70 mbgl, Cl/Br shows that it was contributed by mixing with brackish water. Although SO_4 is not a conservative tracer, its steep downward decline in concentrations is unlikely to reflect only sulphate-reduction given the high concentrations of SO_4 in the shallow aquifers (Fig. 2) and the persistence of SO_4 in brackish-water pockets at depth. Below 70m depth, the lack of SO_4 derived from shallow groundwaters probably also reflects the maximum depth to which they are drawn by pumping, a view consistent with a piston-flow calculation that suggests irrigation pumping has drawn shallow groundwater regionally to depths of ≤ 70 mbgl (Section S5 of the SI). 43,44

Our data confirm the findings of spatial analysis that As-polluted groundwater in Pleistocene aquifers beneath deep palaeo-channels has not been drawn down from *shallow* palaeo-channel aquifers by deep pumping. The As may have been drawn down from overlying As-polluted *deep* palaeo-channel aquifers by deep pumping; or been generated in the Pleistocene grey-sand aquifers close to or at the levels at which it is now found. We examine these two possibilities below.

Stable Isotopes. Downward migration of As-polluted groundwater in deep palaeochannel aquifers to pollute underlying Pleistocene aquifers cannot have happened in historic times because the Cl/Br tracer shows that groundwaters in the Pleistocene aquifers are largely unaffected by contaminant Cl. Such downward movement of groundwater might have happened in pre-historic times before Cl contamination of groundwater became widespread. In this latter case, the groundwaters in deep palaeo-channels and those in the underlying Pleistocene aquifers should have

similar isotopic compositions. Conversely, if the As-bearing groundwater in the deep Pleistocene aquifers is *in-situ*, it should have a different isotopic composition to groundwater in overlying deep palaeo-channels: each type of groundwater would have been recharged at different times under different climatic regimes.

The $\delta^{18}O$ and $\delta^{2}H$ of groundwaters from Holocene deep palaeo-channel aquifers plot along an extended evaporation line that trends to the right of the local meteoric water line (LMWL) of Ref 45 and intersects it at the value for local volume-weighted rainfall (Fig. 4a). Groundwaters from Pleistocene aquifers beneath the deep palaeo-channels cluster tightly around two local evaporation lines (LELs) labelled A and B in Fig. 4a, with slopes different to the LEL for deep palaeo-channel groundwaters. The LELs A and B intercept the LMWL at isotopic compositions more positive than that for annual, volume-weighted, rainfall, which is the intercept on the LMWL of the LEL for deep palaeo-channel groundwater. Deep palaeo-channel groundwaters also have a much wider range of composition than do deeper groundwaters in the Pleistocene aquifers. The majority of groundwaters from beneath palaeo-interfluvial interiors also cluster tightly around LELs A and B, the tightness of fit being particularly close for groundwaters from depths > 150 mbgl.

The isotopic differences between groundwaters in the Holocene deep palaeo-channels and those in the Pleistocene aquifers beneath them (and at depths > 70 mbgl beneath the palaeo-interfluves) shows that the groundwater in the deep Pleistocene aquifers cannot be derived from deep palaeo-channel groundwater. It follows that the As-polluted groundwater in Pleistocene aquifers beneath deep palaeo-channels has obtained its As in or near its present locality and depth. We postulate that it has been generated *in-situ* by microbial reduction of sedimentary iron oxyhydroxides and discuss the potential driver for that reaction in a later section, whilst digressing slightly here to examine the special case of wells at palaeo-interfluvial margins.

Palaeo-interfluvial Margins. Direct recharge downward into palaeo-interfluvial aquifers to replace abstracted groundwater is prevented by the LGMP, so recharge is focused at their margins and occurs from adjacent aquifers, and by upflow from deeper levels (Fig. S2; Refs 31–33). It is therefore at palaeo-interfluvial margins that the penetration to depth of As and Cl from shallow levels should be greatest. Below, we examine the evidence for such penetration in our field area.

Concentrations of As in groundwater from Pleistocene aquifers are lower beneath the palaeo-interfluvial margins than in groundwaters from beneath deep palaeo-channels. Also, the marginal sites show their highest concentrations at slightly shallower depth than are seen at deep palaeo-channel locations (Figs. 2, S3).

Of 22 groundwaters from beneath palaeo-interfluvial margins, only 7 have Cl/Br much above the brackish-water mixing-line (Fig. 3). Of these 3 contain \leq 3 µg/L As, the others contain 36

to 135 μ g/L As. The Cl mixing model between shallow As-polluted groundwater and uncontaminated recharge (Section S6 of the SI) shows that shallow groundwaters contributes ≤ 10 μ g/L to groundwater under palaeo-interfluvial margins, which is $\leq 12\%$ of the As in these wells, and mostly much less. These figures show that few of the As-affected groundwaters in Pleistocene aquifers beneath palaeo-interfluvial margins derive significant As from shallow groundwaters.

The profiles and concentrations of As are consistent with the movement into Pleistocene aquifers at palaeo-interfluvial margins of groundwaters mostly from Pleistocene aquifers *beneath* deep palaeo-channels, with localized contributions from palaeo-channel groundwaters. The widespread presence of the LGMP prevents downward recharge of palaeo-interfluves, thereby focusing flow into them through palaeo-channels.³¹⁻³³ Alluvial channel deposits are known for their heterogeneity and discontinuous fine-grained units (*e.g.* abandoned-channel fill³³) restrict vertical flow more than they restrict horizontal flow. Deep rotary drilling in the Bengal Basin²⁶ tends to confirm this general rule as has our shallower drilling (Fig. S7).

The Redox Driver. We have shown that As is present in groundwater in deep Pleistocene aquifers of the study area, and that it did not migrate to those aquifers from overlying levels. It must therefore be in-situ. How did it arise? We postulate that is arises from reductive dissolution of sedimentary iron oxyhydroxides, a mechanism widely accepted as the cause of natural As-pollution of anoxic groundwaters worldwide. He further postulate that the driver of reduction was dissolved organic matter in Late Pleistocene or earliest Holocene river-water that infiltrated downward through river beds. When induced by pumping to infiltrate river banks, organic matter in river water drives reduction in aquifers 22-54 and natural infiltration of organic matter would have a similar effect. The opportunity for such infiltration would have been greatest between 50 ka and 18 ka as sea-level declined 55, the coastline retreated southwards from the study area, and the climate became gradually drier. The declining sea-level would have left river beds at increasing elevations, a maximum of 125 m above sea level being reached at the LGM at 18 ka.

5. IMPLICATION for MITIGATION

Modelling the migration to depth of As in groundwater of the Bengal Basin^{26,27,59,60} predicts a variety of outcomes from deep pumping. Those outcomes will be modified as local knowledge at large scale refines the assumptions that underpin such models. In the mostly rural settings we have studied, the deep As is relict and unconnected to pumping. Our findings are based on a smallish region of central West Bengal. Nevertheless, palaeo-channels and palaeo-interfluves are found across much of the Bengal Basin^{3,4,19,20,30-33} so our findings will likely apply widely. Further surveys of groundwater arsenic, coupled to analysis of Cl/Br and stable isotopes, ideally with

tritium/¹⁴C analysis as well, are needed to show just how widespread across the Bengal Basin our findings will be applicable.

Arsenic pollution in deep groundwater in West Bengal has been reported before^{24,25} but those authors report a linear decrease in As concentrations with depth, a trend not seen in our work, so care is needed when comparing those studies' results to ours. The different trends with depth may reflect the different well-types sampled. The studies of Refs. 24 and 25 appear to have sampled deep high-yield municipal wells with screens between 10 and 30 m long. The mixed-redox state of many of their groundwaters, which contained reduced and oxidised species (e.g. dissolved O_2 and dissolved Fe) may reflect mixed waters sampled by such long screens. High-yield wells may draw down shallow groundwater locally, as those authors state, but perhaps only to the top of the screen. For this study, we sampled mostly village, hand-pumped, wells of low yield with screen lengths typically ≤ 11 m because we sought to quantify the regional effect of drawdown by irrigation pumping, rather than the localized and specific effect of any particular high-yield well. Given our findings, it is unwise to use as groundwater tracers either Cl without Br, or $\delta^{18}O$ without δ^2H , especially when interpreting depth profiles.

To avoid As-pollution fully, community wells may need to be place remote from major pumping centres. In order to avoid natural deep As, they may also need to be deeper than the 500 ft (152 m) depth specified by the Government of West Bengal as the presumed depth to As-free water. Mitigation through the installation of deep wells would be best accomplished in the light of a detailed knowledge of the subsurface distribution of palaeo-channels and palaeo-interfluves because sedimentological setting is one of the main controls on the risk of As-pollution of the Pleistocene, aquifers.

If screened beneath a palaeo-interfluve and > 50 m from its margin, wells of low-yield that tap Pleistocene aquifers are likely to remain safe for decades. The exception will occur where a low-yield well lies between a high-capacity well and a palaeo-interfluvial margin. Greatest protection will accrue to low-yield wells tapping brown sand beneath palaeo-interfluves at around depths of 50 - 70 mbgl because such sands sorb $As^{27,32,61}$ that may migrate laterally from adjacent aquifers.

Wells that tap Pleistocene aquifers beneath palaeo-channels at depths between 120 and 180 mbgl are liable to encounter natural As-pollution. Local knowledge of that deep arsenic will typically be lacking, so appreciating that regulation operates remotely from local knowledge, wells deeper than 200 m may be prudent for low-yield wells beneath deep palaeo-channels in order to provide reasonable assurance that low-As groundwater can be obtained. The top of the screen should be as far as is practicable below the base of any overlying palaeo-channel in order to minimize the hazard from drawdown of As-polluted groundwater in deep palaeo-channels over the

long term. Annual well-testing should be sufficient to identify encroaching As and could be conducted using field kits which, in competent hands, are sufficiently accurate for such a purpose. We reiterate the view⁶² that the benefits of abstracting groundwater *via* wells that tap the Late Pleistocene aquifer outweigh the risk from migration of As, provided those benefits are not subject to elite capture as described in Ref. 63.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the via the Internet at http://pubs.acs.org.

Detailed sedimentology, methods, draw-down calculations, well-nest profiles, supporting tables, and supporting figures (PDF)

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Notes

The authors declare no competing financial interest.

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REFERENCES

- (1) WHO. Guidelines for Drinking-Water Quality, 4th ed.; Geneva: World Health Organization, 2011.
- (2) PHED 1991. National Drinking Water Mission Project on Arsenic Pollution in Groundwater in West 378 Bengal. Final Report, Steering Committee, Arsenic Investigation Project, PHED, Kolkata, Govt. of West Bengal, India, 1991, 57pp.

- (3) DPHE 1999. Groundwater Studies for Arsenic Contamination in Bangladesh. Phase I: rapid investigation. Department of Public Health Engineering, Government of Bangladesh. Mott MacDonald Ltd (UK) and British Geological Survey (6 vols). 1999.
- (4) DPHE 2001. Arsenic contamination of groundwater in Bangladesh. In: Kinniburgh, D.G., Smedley, P.L., eds.; BGS Technical Report WC/00/19. British Geological Survey, Keyworth (3 Vols.), 2001.
- (5) BBS/UNICEF 2011. Bangladesh national drinking water quality survey 2009. Dhaka: Bangladesh Bureau of Statistics and UNICEF. 2011.
- (6) Chakraborty, D.; Das, B.; Rahman, M. M.; Chowdhury, U. K.; Biswas, B.; Goswami, A. B.; Nayak, B.; Pal, A.; Sengupta, M. K.; Ahamed, S.; Hossain, A.; Basu, G.; Roychowdhury, T.; Das, D. Status of groundwater arsenic contamination in the state of West Bengal, India: a 20-year study report. *Mol. Nutr. Food Res.* 2009, 53, 542 551.
- (7) Chatterjee, A.; Das, D.; Mandal, B. K.; Roy Chowdhury, T.; Samanta, G.; Chakraborti, D. Arsenic in groundwater in 6 districts of West Bengal India: the biggest arsenic calamity in the world, Part 1. Arsenic species in drinking water and urine of the affected people. *Analyst*, 1995, 120, 645–50.
- (8) Dhar, R. K.; Biswas, B. K.; Samanta, G.; Mandal, B. K.; Chakraborti, D.; Roy, S. et al. Groundwater arsenic calamity in Bangladesh. *Current Science* 1997, 73, 48–59.
- (9) Smith, A. H.; Lingas, E. O.; Rahman, M. Contamination of drinking-water by arsenic in Bangladesh: a public health emergency. *Bull. World Health Organ.* 2000, 78, 1093–1103.
- (10) Chakraborti, D.; Sengupta, M. K.; Rahman, M.N.; Chowdhury, U. K.; Lodh, D.; Chanda, C.R.; Basu, G. K.; Mukherjee, S.C.; Saha, K. C. Groundwater arsenic exposure in India, In: Chappell, W. R., Abernathy, C. O., Calderon, R. L., Thomas, D. J., Eds.; Arsenic Exposure and Health Effects (V). Elsevier. 2003, 3–24.
- (11) Yu, W. H.; Harvey, C. M.; Harvey, C. F. Arsenic in groundwater in Bangladesh: a geo-statistical and epidemiological framework for evaluating health effects, and potential remedies. *Water Res. Research*, 2003, 39, 1146–1163.
- (12) Flanagan, S. V.; Johnston, R. B.; Zheng, Y. Arsenic in tube well water in Bangladesh: health and economic impacts and implications for arsenic mitigation. *Bulletin of the World Health Organization* 2012, 90, 839–846, doi: 10.2471/BLT.11.101253.
- (13) Johnston, R.; Hug, S. J.; Inauen, J.; Khan, N. I.; Mosler, H.-J.; Yang, H. Enhancing arsenic mitigation in Bangladesh: findings from institutional, psychological, and technical investigations. *Sci. Total Environ.* 2014, 488–489, 477–483.

- (14) Ravenscroft, P.; McArthur, J. M.; Hoque, B. Geochemical and palaeohydrological controls on pollution of groundwater by arsenic. Arsenic Exposure and Health Effects IV. Chappell, W.R., Abernathy, C.O., Calderon R. Eds.; Elsevier Science Ltd., Oxford. 2001, 53–77.
- (15) van Geen, A.; Ahmed, K. M.; Seddique, A. A.; Shamsudduha, M. Community wells to mitigate the arsenic crisis in Bangladesh. *Bull. World Health Org.*, 2003, 81, 632–638.
- (16) van Geen, A.; Cheng, Z.; Jia, Q.; Seddique, A. A.; Rahman, M. W.; Rahman, M. M.; Ahmed, K. Z. Monitoring 51 community wells in Araihazar, Bangladesh, for up to 5 years: implications for arsenic mitigation. *J. Environ. Sci. Health Part A*, 2007, 42, 1729–1740.
- (17) Zheng, Y.; van Geen, A.; Stute, M.; Dhar, R.; Mo, Z.; Cheng, Z.; Horneman, A.; Gavrieli, I.; Simpson, H.J.; Versteeg, R.; Steckler, Grazioli-Venier, A.; Goodbred, S.; Shahnewaz, M.; Shamsudduha, M.; Hoque, M.A.; Ahmed K.M. Geochemical and hydrogeological contrasts between shallow and deeper aquifers in two villages of Araihazar, Bangladesh: Implications for deeper aquifers as drinking water sources. *Geochim. Cosmochim. Acta*, 2005, 69, 5203–5218. Doi: 10.1016/j.gca.2005.06.001.
- (18) von Brömssen, M.; Jakariya, M.; Bhattacharya, P.; Ahmed, K. M.; Hasan, M. A.; Sracek, O.; Jonsson, L.; Lundell, L.; Jacks, G. Targeting low-arsenic aquifers in Matlab Upazila, Southeastern Bangladesh. *Sci. Total Environ.* 2007, 379, 121–132.
- (19) Hoque, M. A.; McArthur, J. M.; Sikdar, P. K. The palaeosol model of arsenic pollution of groundwater tested along a 32 km traverse across West Bengal, India. *Sci. Total Environ*. 2012,431,157–165.
- (20) Hoque, M. A.; McArthur, J. M.; Sikdar, P. K. 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 *Hydrogeology Journal*. 2014; doi 10.1007/s10040-014-1139-8.
- (21) GoB 2004. National policy on arsenic mitigation, and its implementation plan. Government of Bangladesh; 2004.
- (22) Ravenscroft, R.; Kabir, A.; Hakim, S. A. I.; Ibrahim, A. K. M.; Ghosh, S. K.; Rahman, Md. S.; Akhter, F.; Sattar, Md. A. Effectiveness of public rural waterpoints in Bangladesh with special reference to arsenic mitigation. *Jour. Water, Sanitat. Hygiene Develop.*. 2014, 4, 545–562.
- (23) Aggarwal, P. K.; Basu, A. R.; Poreda, R. J. Preliminary Report of Investigations. Isotope Hydrology of Groundwater in Bangladesh: Implications for Characterization and Mitigation of Arsenic in Groundwater IAEA TC project (BGD/8/016), Vienna, 2000.
- (24) Mukherjee, A.; Fryar, A. E. Deeper groundwater chemistry and geochemical modeling of the arsenic affected western Bengal basin, West Bengal, India. *Appl. Geochem.* 2008, 23, 863–894.

- (25) Mukherjee, A.; Fryar, A. E.; Scanlon, B. R.; Bhattacharya, P.; Bhattacharya, A. Elevated arsenic in deeper groundwater of the western Bengal Basin, India: extent and controls from regional to local scale. *Appl. Geochem.* 2011, 26, 600–613.
- (26) Burgess, W. G; Hoque M. A.; Michael, H. A; Voss, C. I.; Breit, G. N.; Ahmed K. M. Vulnerability of deep groundwater in the Bengal aquifer system to contamination by arsenic. *Nat. Geosci.* 2010, 3, 83–87.
- (27) Radloff, K. A.; Zheng, Y.; Michael, H. A.; Stute, M.; Bostick, B. C.; Mihajlov, I.; Bounds, M.; Huq M. R.; Choudhury I.; Rahman M.W.; Schlosser P.; Ahmed K. M.; van Geen A. Arsenic migration to deep groundwater in Bangladesh influenced by adsorption and water demand. *Nat. Geosci.* 2011, 4,793–8.
- (28) Hug, S. J.; Gaertner, D.; Roberts, L. C.; Schirmer, M.; Ruettimann, T.; Rosenberg, T. M.; Badruzzaman, A. B. M.; Ashraf Ali, M. Avoiding high concentrations of arsenic, manganese and salinity in deep tubewells in Munshiganj District, Bangladesh. *App. Geochem.* 2011, 26, 1077–1085.
- (29) CGWB 2011. Report on the dynamic ground water resources of West Bengal as on 31.03.2009. Central Ground Water Board and State Water Investigation Directorate, Technical Report Series 'B', 2011, 234, 1-244.
- (30) Ghosal, U.; Sikdar, P. K.; McArthur, J.M. Palaeosol control of arsenic pollution in deltaic aquifers: the example of West Bengal, India. *Groundwater*. 2014, doi: 10.1111/gwat.12251.
- (31) McArthur, J. M.; Ravenscroft, P.; Banerjee, D. M.; Milsom, J.; Hudson-Edwards, K. A.; Sengupta, S.; Bristow, C.; Sarkar, A.; Tonkin, S.; Purohit, R. How paleosols influence groundwater flow and arsenic pollution: A model from the Bengal Basin and its worldwide implication. *Water Resour. Res.* 2008, 44, W11411, doi: 10.1029/2007WR006552.
- (32) McArthur, J. M.; Sikdar, P. K.; Nath, B.; Grassineau, N.; Marshall, J.D.; Banerjee D. M. Sedimentological control on Mn, and other trace elements, in groundwater of the Bengal Delta. *Environ. Sci. Technol.* 2012a, 46, 669–676.
- (33) Desbarats, A. J.; Koenig, C. E. M.; Pal, T.; Mukherjee, P. K.; Beckie, R. D. Groundwater flow dynamics and arsenic source characterization in an aquifer system of West Bengal, India. *Water Resour. Res.* 2014, 50, 4974–5002, doi: 10.1002/2013WR014034.
- (34) Karim, M.; Komori, Y.; Alam, M. Arsenic occurrence and depth of contamination in Bangladesh. *Jour. Environ. Chem.* 1997, 7, 783–792.
- (35) Acharyya, S. K.; Chakraborty, P.; Lahiri S.; Raymahashay B. C.; Guha, S.; Bhowmik, A. Arsenic poisoning in the Ganges delta. *Nature*, 1998, 401, 545.

- (36) Chowdhury, T. R.; Basu G. K.; Mandal, B. K.; Biswas, B. K.; Samanta, G.; Chowdhury, U. K.; Chanda, C. R.; Lodh, D.; Roy S. L.; Saha K. C.; Roy S.; Kabir, S.; Quamruzzaman, Q.; Chakraborti, D. Arsenic poisoning in the Ganges delta. *Nature*, 1998, 401, 545–546.
- (37) Davis, S. N.; Whittemore, D. O.; Fabryka-Martin, J. Uses of chloride/bromide ratios in studies of potable water. *Ground Water*. 1998, 36, 338–350.
- (38) Vengosh, A.; Pankratov, I. Chloride/bromide and chloride/fluoride ratios of domestic sewage effluents and associated contaminated ground water. *Ground Water*, 1998, 36, 815–824.
- (39) Panno, S. V.; Hackley, K. C.; Hwang, H. H.; Greenberg, S. E.; Krapac, I. G.; Landsberger, S.; O'Kelly, D. J. Characterization and identification of Na-Cl sources in ground water. *Ground Water.*, 2006, 44, 176–187.
- (40) McArthur, J. M.; Sikdar, P. K.; Hoque, M. A.; Ghosal, U. Waste-water impacts on groundwater: Cl/Br ratios and implications for arsenic pollution of groundwater in the Bengal Basin. *Sci. Total Environ.* 2012b, 437, 390–402.
- (41) Lowers, H. A.; Breit, G. N.; Foster, A. L.; Whitney, J.; Yount, J.; Uddin, M. N.; Muneem, A.A. Arsenic incorporation into authigenic pyrite, Bengal Basin sediment, Bangladesh. *Geochim Cosmochim Acta*; 2007, 71, 2699–717.
- (42) Nissenbaum, A.; Magaritz, M. Bromine-rich groundwater in the Hula Valley, Israel. *Naturwissenschaften* 1991, 78, 217–218.
- (43) Harvey, C. F.; Swartz, C. H.; Badruzzaman, A. B. M.; Keon-Blute, N.; Yu, W.; Ali, M. A.; Jay, J.; Beckie, R.; Niedan, V.; Brabander, D.; Oates, P. M.; Ashfaque, K. N.; Islam, S.; Hemond, H. F.; Ahmed, M. F. Arsenic mobility and groundwater extraction in Bangladesh. *Science* 2002, 298, 1602-1606.
- (44) Klump, S., Kipfer, R., Cirpka, O.A., Harvey, C.F., Brennwald, M.S., Ashfaque, K.N., Badruzzaman, A.B.M., Hug, S.J., Imboden, D.M., 2006. Groundwater dynamics and arsenic mobilization in Bangladesh assessed using noble gases and tritium. *Environ. Sci. Technol.* 40, 243e250.
- (45) Sengupta, S.; Sarkar, A. Stable isotope evidence of dual (Arabian Sea and Bay of Bengal) vapour sources in monsoonal precipitation over north India. *Earth Planet. Sci. Lett.* 2006, 250, 511–521.
- (46) Korte, N.E.; Fernando, Q. A review of arsenic(III) in groundwater. *Crit. Rev. In Environm. Control*, 1991, 21, 1–39.
- (47) Matisoff, G.; Khourey, C.J.; Hall, J.F.; Varnes, A.W.; Strain, W. The nature and source of arsenic in Northeastern Ohio ground water. *Ground Water* 1992, 20, 446–455.
- (48) Nickson, R.; McArthur, J.; Burgess, W.; Ahmed, M.; Ravenscroft P.; Rahman, M. Arsenic poisoning of groundwater in Bangladesh. *Nature*, 1998, 395, 338.

- (49) Ravenscroft, P.; Brammer, H.; Richards, K. S. Arsenic Pollution: A Global Synthesis Wiley-Blackwell. 2009, 588pp.
- (50) Fendorf, S.; Michael, H.A.; van Geen A. Spatial and temporal variations of groundwater arsenic in South and Southeast Asia. *Science* 2010, 328, 1123–1127.
- (51) Guo, H.; Wen, D.; Liu Z.; Jia, Y.; Guo Q. A review of high arsenic groundwater in Mainland and Taiwan, China: distribution, characteristics and geochemical processes. *Appl. Geochem.* 2014, 41, 196–217.
- (52) Bourg, A. C. M.; Bertin, C. Biogeochemical processes during the infiltration of river water into an alluvial aquifer. *Environ.Sci. Technol.*, 1993, 27, 661–666.
- (53) Farnsworth, C. E.; Hering, J.G. Inorganic geochemistry and redox dynamics in bank filtration settings. *Environ. Sci. Technol.* 2011, 45, 5079–5087.
- (54) van Geen, A.; Bostick, B.C.; Trang, P. T. K.; Lan V. N.; Mai, N-N.; Manh, P. D.; Viet, P. H.; Radloff K.; Aziz Z.; Mey J. L.; Stahl M. O.; Harvey C. F.; Oates P.; Weinman B.; Stengel C.; Frei F.; Kipfer R.; Berg M. Retardation of arsenic transport through a Pleistocene aquifer. *Nature*, 2013, 501, 204–208.
- (55) Lambeck K.; Esat, T.M.; Potter, E.-K. Links between climate and sea levels for the past three million years. *Nature*, 419, 199–206.
- (56) Herzschuh, U. Palaeo-moisture evolution in monsoonal Central Asia during the last 50,000 years. *Quaternary Science Reviews*, 2006, 25, 163–178.
- (57) Chen X.; Chen, F.; Zhou, A.; Huang, X.; Tang, L.; Wu, D.; Zhang, X.; Yu, J. Vegetation history, climatic changes and Indian summer monsoon evolution during the Last Glaciation (36,400–13,400 cal yr BP) documented by sediments from Xingyun Lake, Yunnan, China. *Palaeogeog. Palaeoclimatol. Palaeoecol*, 2014, 410, 179–189
- (58) Evans, D.; Bhatia, R.; Stoll, H.; Müller, W. LA-ICP-MS Ba/Ca analyses of planktic foraminifera from the Bay of Bengal: implications for late Pleistocene orbital control on monsoon freshwater flux. *Geochemistry, Geophysics, Geosystems*. 2015. Doi: 10.1002/2015GC005822
- (59) Michael, H. A.; Voss, C. I. Evaluation of the sustainability of deep groundwater as an arsenic-safe resource in the Bengal Basin. *Proc. Natl. Acad. Sci.* 2008, 105, 8531–8536.
- (60) Michael, H. A.; Voss, C. I. Controls on groundwater flow in the Bengal Basin of India and Bangladesh: regional modeling analysis. *Hydrogeol Jour.* 2009, 17, 1561–1577.
- (61) McArthur, J. M.; Banerjee, D. M.; Sengupta, S.; Ravenscroft, P.; Klump, S.; Sarkar, A.; Disch, B.; Kipfer R. Migration of As, and 3H/3He ages, in groundwater from West Bengal: implications for monitoring. *Water Res.* 2010, 44, 4171–4185.

- (62) Ravenscroft, P.; McArthur, J. M.; Hoque, M. Stable groundwater quality in deep aquifers of southern Bangladesh: the case against sustainable abstraction. *Sci. Tot. Environ.*, 2013, 454–455, 627–638.
- van Geen, A.; Ahmed, K. M.; Ahmed, A. B.; Choudhury, I.; Mozumder, M. R.; Bostick B. C.; and Mailloux B. J. Inequitable allocation of deep community wells fpr reducing arsenic exposure in Bangladesh. Jour. Water, San. Hyg. Delevelop. In press, 2015.

List of Figures

- Fig. 1. a) Study area, location of sampled wells, all > 70 m deep, and aquifer types. Areas shaded yellow are subsurface palaeo-interfluves with tops 35 38 mbgl; deep palaeo-channels exist under unshaded areas (modified from Ref. 30). Cross-section along line A-B is shown in Fig. S1 (Supporting Information), with the solid part of the line schematized in Fig. S2. b). Spatial distribution of As in groundwaters, all of which are from depths > 70 m.
- Fig. 2. Depth profiles of As and SO_4 in groundwater. Note scale changes on concentration axes. Data for depths < 70 m are from Ghosal et al. (2015). Concentrations of As beneath deep palaeochannels form a broad maxima between 120 and 180 mbgl.
- Fig. 3. Relation of Cl concentration to Cl/Br in groundwaters from depths > 70 m. Of 22 groundwaters from beneath the margins of palaeo-interfluves (dotted symbols), 7 fall much above the brackish mixing line. End-member compositions for mixing lines from Ref 40 as follows: uncontaminated recharge, 1.3 mg/L Cl, 9.5 μ g/L Br; waste-water, 126 mg/L Cl, 81 μ g/L Br; brackish connate water (10% seawater), 1,940 mg/L Cl, 6.8 mg/L Br. Analytical uncertainty of Cl/Br values is about the size of the symbols.
- Fig. 4. a) Stable-isotopic composition of groundwaters from deep palaeo-channels and from Pleistocene aquifers beneath deep palaeo-channels. With one exception labelled SBN 11, data for Pleistocene groundwaters cluster tightly around LELs A and B, which are reduced-major-axis regression fits to the local data pools. Data for deep palaeo-channels scatter widely along the LEL for deep palaeochannel groundwaters, the LEL has a slope steeper than LEL A or LEL B, and a lower intercept on the LMWL. The local meteoric water line (LMWL) from Ref. 45 is for Barasat, 50 km south of the study area. The large yellow triangle is those authors' value of annual volume-weighted rainfall for Barasat of -6.20 for δ^{18} O and -39.9 for δ^{2} H. b) Stable isotopic composition of groundwaters from Pleistocene aquifers beneath palaeo-interfluvial interiors of the study area. These also tightly group around LEL A and B, especially for groundwaters deeper than 150 mbgl.

Fig. 1

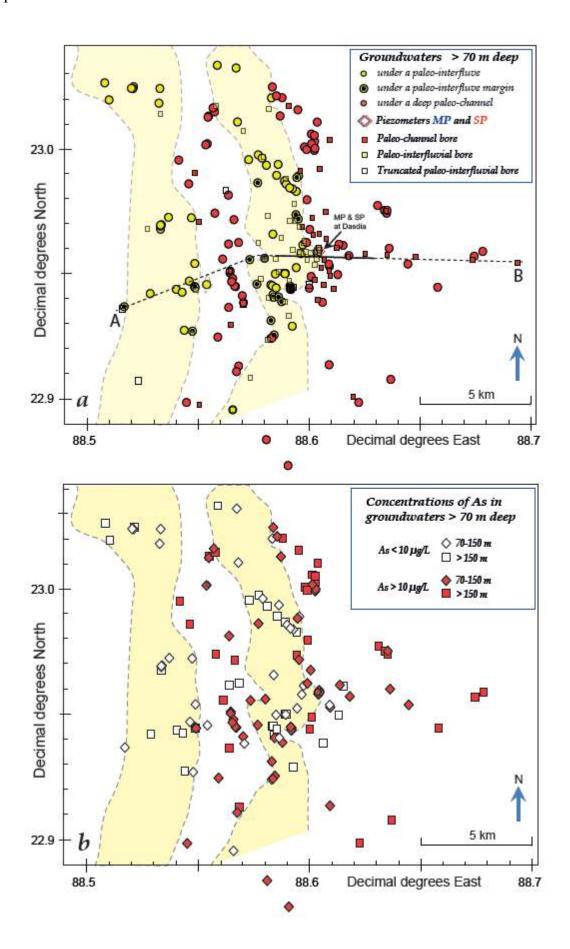


Fig. 2.

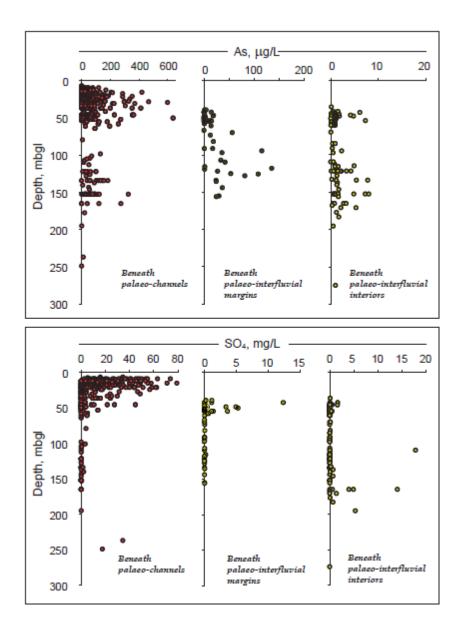


Fig. 3.

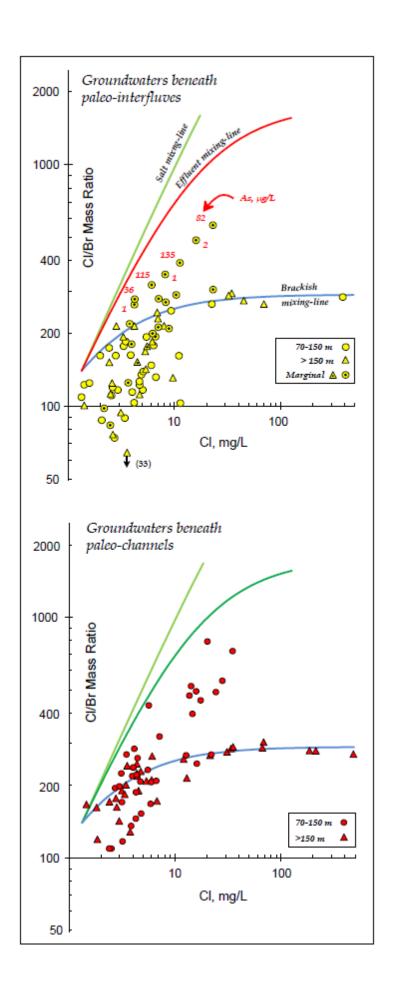
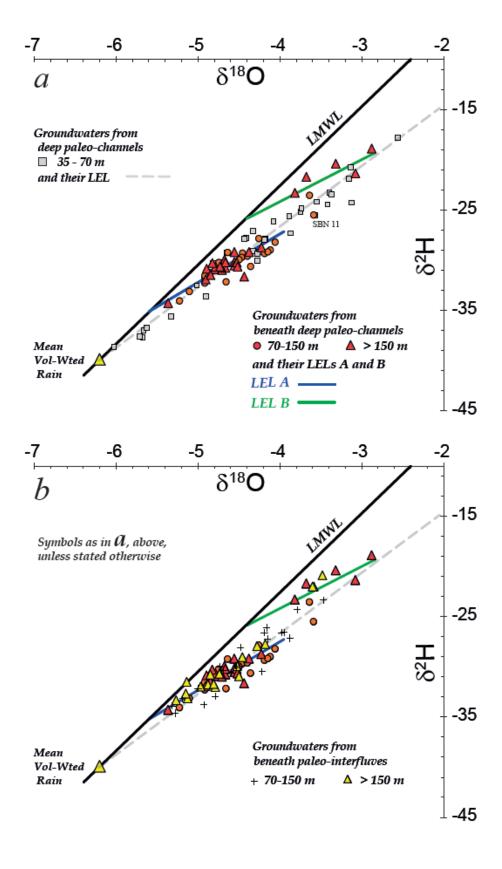


Fig. 4.



Supplementary Information

Arsenic in groundwater: the deep Late Pleistocene aquifers of the western Bengal Basin.

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17 pages including Table S1 6 figures 1 table

S1. History of the Bengal Basin, 125 ka to Now. The sedimentology of the aquifers of the Bengal Basin is summarized from Refs. 1 – 9. In outline, sea level declined by some 120 m between 125 ka and 18 ka, the time of the last glacial maximum (LGM)¹⁰. During the decline, the coastline retreated southward by up to 100 km^{5,6,11}. The existing and newly-exposed coast, and the low-lying hinterland, was eroded into a landscape of interfluves and river channels with a relief of mostly no more than 50 m^{3,4,8}. The pre-LGM sediments, predominantly sands, beneath the interfluves were weathered in the zone of most active groundwater flow, largely the unsaturated zone down to depths level with river bases, which was generally no more than 30 m depth. The weathering formed the oxidised brown Pleistocene sands that now sit on underlying grey Pleistocene sands that were weathered much less. A reddish-brown clay palaeosol formed on top of the brown, weathered sand, and has been termed a laterite^{3,4} and the 'last glacial maximum palaeosol', or LGMP¹².

Sea level rose after the termination of the LGM, stabilising around 6 ka close to the present level. The rise created the accommodation space for the accumulation of post-LGM sediments, largely sands, on top of the LGM landscape to form the present submarine and subaerial deltas, under which lies the buried landscape of palaeo-interfluves and palaeo-channels. River avulsion since 6 ka has reworked the upper 20 - 30 m of the sequence, leaving large areas covered by shallow palaeo-channel fill of grey sands to 20 - 30 m depth, interspersed with silty/clayey/peaty floodplain sequences^{8,13-15}. Historical floodplain silts now cover the succession in the basin's south and central parts, forming an upper, semi-confining aquitard of fine sediments usually ≤ 5 m thick.

Across the Bengal Basin, the LGM land-surface that separates pre- and post-LGM aquifers lies at a depth that depends on location. It has been customary to separate the 'deep' aquifers from the 'shallow' aquifers of the Bengal Basin using a practical division based on a maximum sea-level lowering of 120 m. In the light of both old¹ and recent^{9,14,16} results on sedimentology and dating in the Bengal Basin, that figure is no longer tenable. For example, around the basin margin, the pre-LGM surface crops out, as do pre-LGM aquifers. Additionally, across much of the basin interior, the LGM surface lies at 30–40 m depth where palaeo-interfluves are preserved in the subsurface, and at depths between 50 and 120 m depth in the far smaller areas underlain by palaeo-channels. ^{1-4,8,8,12-15,17-21}

At the LGM, the depth of river incision, relative to today's ground surface, would have been greatest at the coast and been close to the sea-level lowering of around 120 m below present sea-level, which would be around 130 m below present ground level in our area where elevations are currently no more than 10 m above sea-level. Our area was >100 km from the coast at that time^{5,6,11} so the depth of river incision in our field area would have been less than 120 m and probably no more than 70 mbgl on the basis of our drilling information¹⁵ and inferences from the cross-sections of Refs. 3 and 4. For an area some 50 km south of our study area, a depth to the LGM surface¹⁵ is around 50 m. For central

West Bengal, the depth to the LGM surface²² is around 60 m. Using our more relevant local field data, in this work we have assumed that the depth to the LGM surface is 70 mbgl. Our conclusions would not differ were we to set the LGM surface deeper at, say, 100 m, because few of the wells we have sampled have depths between 70 and 100 m bgl depth.

S2. Geology of the Field Area. Reproduced below (Fig. S1) from Ref. 15 is a cross section through the field area from West to East. The line of section is line A-B of Fig. 1.

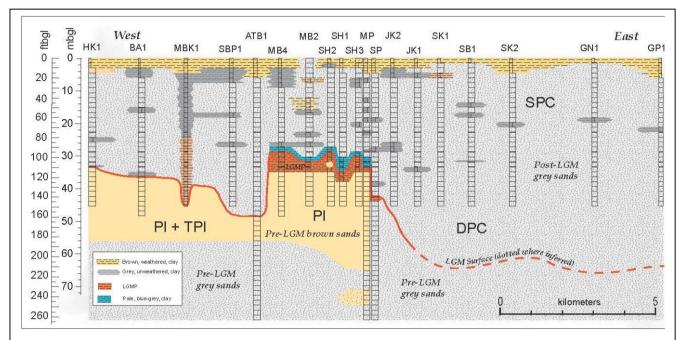


Fig. S1 Cross-section along line A-B in Fig. 1. LGMP = last glacial maximum palaeosol, which is overlain in some locations by a pale, blue-grey, clay. PI = palaeo-interfluve. DPC = deep palaeo-channel. SPC = shallow palaeo-channel. TPI = truncated palaeo-interfluve, where river incision since 6 ka has cut downwards and through the LGMP that caps the palaeo-interfluves.

The eastern two-thirds of the profile have been schematised in Fig. S2 in order to illustrate the hypothetical vulnerability of wells in Pleistocene aquifers to downward movement of As-pollution in overlying Holocene aquifers. Groundwaters in Pleistocene aquifers beneath deep palaeo-channels

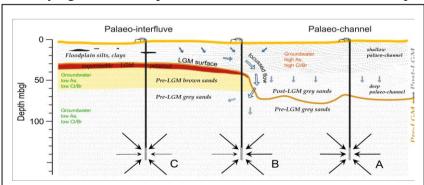


Fig. S2. Schematic cross-section of the solid part of the line of profile A–B in Fig. 1 showing conceptualised command areas of wells in three settings: A, under a deep palaeo-channel. B, under a palaeo-interfluvial margin. C, under a palaeo-interfluvial interior. The well at B draws groundwater from beneath both palaeo-interfluves and deep palaeo-channels.

(Well A in Fig. S2) are more at risk of As-pollution from this source than are those beneath palaeo-interfluves (Well C in Fig. S2). The highest risk occurs at palaeo-interfluvial margins (Well B in Fig. S2) where downward flow is focused because most recharge to palaeo-interfluvial aquifers derives from adjacent aquifers, a lesser amount deriving from upflow beneath the palaeo-interfluves.

S3. Analytical Methods. Samples from wells and piezometers were

taken after purging into two 15 ml polythene tubes, one acidified in the field with 0.15 ml of 50%

Analar® nitric acid for cation analysis, one unacidified for anion analysis. Samples were filtered using 0.45 micron membrane filters only when visibly turbid because we, and others²³, have found that filtered and unfiltered analysis is indistinguishable unless turbid, and because we wished to measure the As concentration in water consumed, which is not filtered after being drawn. Chemical analysis was by ion chromatography for Cl and SO₄ and ICP-MS for As and Br on acidified samples. For ICP-MS, standards were matrix matched to an average groundwater composition for groundwater from West Bengal. Bromide standards were used and mass 79 measured. Backgrounds were high but very stable at around 2,000 cps from peak-spread on the Ar dimer at mass 80, enabling an RSD of $\leq 10\%$ RSD down to 10 µg/L Br, and \pm 1 µg/L Br below that concentration. Sensitivity was around 500 cps/µg/L Br; the detection limit was around 1 µg/L Br. Analysis of oxygen and hydrogen stable isotopes were done on unacidified samples, filtered through 0.22 µm membrane filters, using a Picarro WSCRDS laser instrument calibrated against standards USGS 46 and 48, with USGS 47 run as a sample to provide a further check on data quality. Analytical precision was \pm 0.13‰ for δ^{18} O and \pm 0.45 for δ^{2} H (2s.d.).

S4. Composition of Groundwaters from Well-Nests SP and MP

Our interpretation is that As in deep groundwaters in Pleistocene aquifers has not been drawn down from shallow aquifers, even at palaeo-interfluvial margins. This contention receives support from the vertical distribution of As, Cl, and Cl/Br in two piezometers, MP and SP, located within the

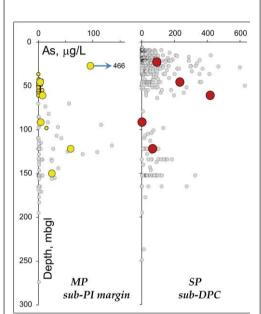


Fig. S3. Depth profile of As in piezometers SP, at a palaeo-interfluvial margin, and MP, located 120 m to the west of SP towards the palaeo-interfluvial interior. Small grey symbols represent other wells sampled in this study.

site SP is natural and essentially *in-situ*. We postulate that the As at depth at site MP has been drawn-in laterally under the palaeo-interfluvial margin by pumping and derives largely from the Pleistocene aquifer underlying the adjacent deep palaeo-channel aquifer, with no contribution from shallow levels and a minimal contribution from the adjacent deep palaeo-channel. A cross-plot of compositions of groundwater from 122 mbgl

field area (Fig. 1). The well-nest SP is located at the edge of a palaeo-channel: two boreholes 10 m apart revealed thin LGMP in the western-most and only grey sand (no LGMP) in the eastern-most. The well-nest MP is 120 m westward of SP in the direction of the interior of the palaeo-interfluve.

The vertical distribution of As at MP and SP (Fig. S3) show high As at shallow depth that is typical of Aspolluted shallow palaeo-channel groundwater. At and below 90 metres depth, Cl concentrations do not exceed 4 mg/L and groundwater Cl/Br is on or below the brackish mixingline of Fig. 3, showing that at these depths, no shallow groundwater is present. Concentrations of As in both piezometers show a minimum at 90 m depth and are higher at deeper levels, a depth distribution also seen in the groundwaters (Fig. 2). This deep As is present in both MP and SP, albeit in concentrations of < 70 μ g/L at depth. We interpret these profiles as showing that the As at depth at

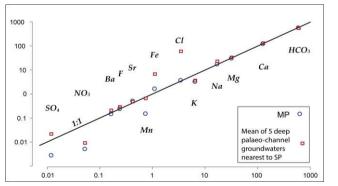


Fig. S4. Compositions of groundwaters at 122 mbgl from piezometer SP compared top that from 122 mbgl from MP and from deep palaeo-channels (35–70 mbgl) nearest to SP. In SP, concentrations of Cl and Fe are close to those in MP but different from those in nearby palaeo-channel groundwaters.

at sites SP and MP show a high degree of concordance (Fig. S4), especially for the more conservative species, supporting the hypothesis of mostly horizontal migration. The concentrations of As at depth differs little between the sites. The uppermost 30 m of the sediments comprising the palaeo-interfluvial aquifers consists of brown sand that can sorb As^{24,25}. At greater depths, the sand is not oxidised, is grey, and presumably contains less iron-oxyhydroxide than the brown sands. Such grey sands will retard As migration less, and possibly hardly at all.

S5. Downward Piston Flow. Drawdown occurs to replace groundwater pumped for domestic supply and irrigation. To combat As-pollution in shallow aquifers, the government of West Bengal installed at least one deep community well per village to a depth of 152 m (500 feet bgl), the abstraction from which is < 10 % of irrigation abstraction. To develop irrigation, from the early 1960's the Government of West Bengal installed numerous high-capacity deep wells for irrigation. These typically are of a total depth between 152 and 190 mbgl, with 176 m being a common depth. Screens are long, typically 30 m. Each well typically supplies around 0.4 km² of Boro (winter) rice with irrigation water during the dry season (February to May) across 74 % of Nadia District and 67% of North 24 Paraganas District. There are no regulations that are effective in restricting well-spacing.

The total depth of flooding by irrigation pumping each year is typically 1 m; we assume that 50% infiltrates and 50% evaporates. Coupled to an aquifer porosity of 40% for unconsolidated sands, these figures equate to total piston-flow drawdown that has penetrated no more than 70 m across Nadia and South 24 Parganas since 1960.

Many factors influence such estimates. Estimates of drawdown will be lessened because not all deep wells are operational at any one time; many farmers prefer to use their own motorised shallow irrigation wells, which pull water from shallower depths; flood irrigation from high-capacity government wells is not applied uniformly, being used only for low ground where hydraulic gradients allow gravity flow to feed fields. Both high-yield irrigation wells and municipal high-yield supply wells have long screens, typically up to 30 m, and the flow of water at any point through the screen will be inversely proportional to its distance from the pump intake: most pumped water comes from the screen area close to the pump intake and little originates from the lowest part of the screen. Drawdown of groundwater in marginal regions of the palaeo-interfluves may be underestimated because recharge to aquifers beneath palaeo-interfluves is focused at such margins.

Migration of groundwater and As from depth to beneath the palaeo-interfluvial aquifer must be occurring in response to extraction from beneath the impermeable LGMP. Drilling shows that the base of the brown sand in the vicinity of the piezometers is at a depth of about 70 mbgl¹⁵, so it is important to note that the deep As has not passed through brown sands beneath the local palaeo-interfluve but has migrated through the underlying grey, pre-LGM, sands. We know of no data that quantifies sorption of As to pre-LGM grey sands, but speculate that such sands sorb less than do brown, palaeo-interfluvial sands, which are known to retard migration of dissolved As in the subsurface of the Bengal Basin^{24–25}.

S6. Cl mixing-models. A few groundwaters from Pleistocene aquifers beneath deep palaeochannels, and a few in Pleistocene aquifers beneath palaeo-interfluvial margins, fall above the Cl v Cl/Br mixing line for brackish water, are contaminated by shallow Cl, and so might be polluted by

shallow As. The proportion of contaminant shallow groundwater, derived from a Cl-based mixing model, 1.4% and 27% for the affected ranges from groundwaters from beneath palaeo-channels, and from 2.3% to 17% for the affected groundwaters from palaeo-interfluvial margins. beneath The concentrations in groundwater in deep palaeo-channels (35 - 70 mbgl) ranges up to 640 µg/L, but 75% of concentrations are < 100 µg/L¹⁵. Groundwater flow contaminant concentrations homogenises hydrodynamic dispersion, so concentrations in any deep palaeo-channel groundwaters that moves downward would likely tend toward the mean or median concentrations, which are, respectively, 60 and 27 ug/L (n = 75; Ref. 15). The maximum concentration of As that groundwater in deep palaeo-channels could supply to groundwaters beneath them would thus be 16 µg/L of As, and for most of the samples discussed would be a good deal less. With groundwaters beneath deep palaeochannels containing up to 326 µg/L As, but mostly concentrations in the range 10 to 100 µg/L, the contribution to deep As from shallow groundwater is likely to be insignificant even in wells with elevated Cl/Br. Even in cases where shallow, high Cl, high Cl/Br, groundwater might migrate to depth, its high content of NO₃ and SO₄ is likely to suppress Aspollution²⁶ by preferential utilisation of NO₃ for oxidation of DOC, and by sequestration of As in neoformed pyrite from sulphate reduction.

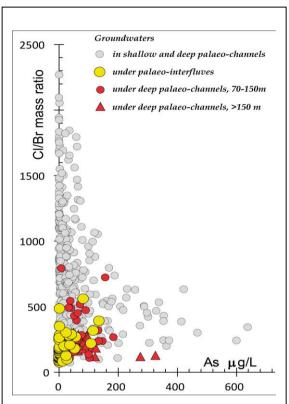
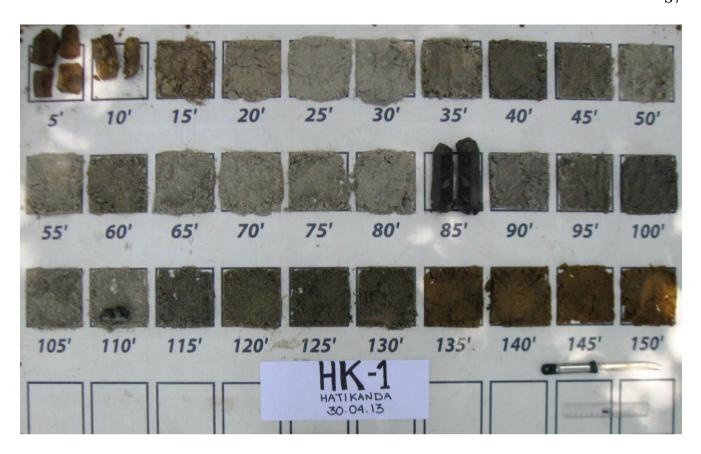


Fig. S5. Relation of As concentrations to Cl/Br mass ratio. The inverse relationship arises through the addition to groundwater of waste water containing high concentrations of Cl and NO₃. The NO₃ suppresses reduction of FeOOH and so prevents reductive dissolution releasing As to groundwater. Data for groundwaters at depths < 70m from Ref. 15.

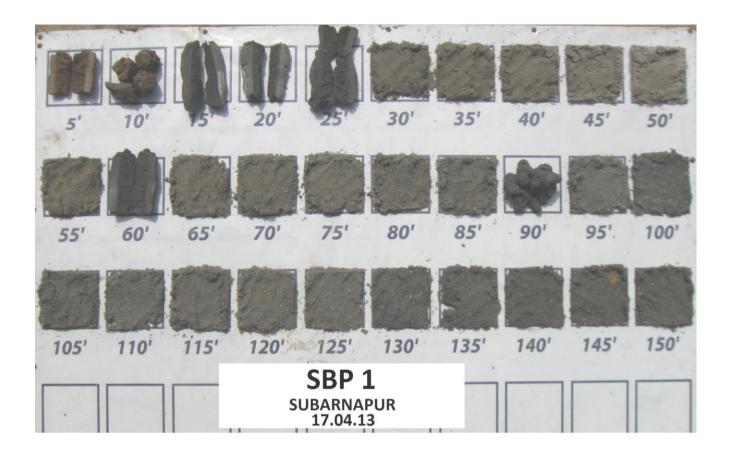
S7. Fig. 6. Photos of Cores

The photos below are a representative selection showing the frequency and nature of clayey lenses in shallow and deep palaeo-channel sediments. For locations, see Ref 15.











References

- (1) Umitsu, M. Late Quaternary sedimentary environments and landforms in the Ganges Delta. *Sed. Geol.* 1993, 83, 177–186.
- (2) DPHE 1999. Groundwater studies for Arsenic contamination in Bangladesh. Phase I: Rapid Investigation., Department of Public Health Engineering (DPHE) of Government of Bangladesh, British Geological Survey (BGS) and Mott MacDonald Ltd (MML) UK.
- (3) Goodbred, S. L.; Kuehl, S. A. The significance of large sediment supply, active tectonism, and eustasy of margin sequence development: Late Quaternary stratigraphy and evolution of the GangesBrahmaputra delta. *Sed. Geol.* 2000, 133, 227–248.
- (4) Goodbred, S. L. Jr.; Kuehl, S. A. Enormous Ganges-Brahmaputra sediment supply, active tectonism, and eustasy on margin sequence development: Late Quaternary stratigraphy and evolution of the Ganges-Brahmaputra delta. *Geology*, 2000, 28, 1083–1086.
- (5) Allison, M. A. Geologic framework and environmental status of the Ganges-Brahmaputra Delta. *Jour. Coastal Res.* 1998,14, 3, 826–836.
- (6) Allison, M. A. Historical changes in the Ganges-Brahmaputra Delta Front. *Jour. Coastal Res.*, 1997, 14, 1269–1275.
- (7) Allison, M. A.; Khan, S. R.; Goodbred S. L.; Kuehl S. A. Stratigraphic evolution of the late Holocene Ganges–Brahmaputra lower delta plain. *Sed. Geol.* 2003, 155, 317–342
- (8) Goodbred, S. L. Jr.; Kuehl, S.A.; Steckler, M. S.; Sarkar, M. H. Controls on facies distribution and stratigraphic preservation in the Ganges-Brahmaputra delta sequence. *Sed. Geol.* 2003,155, 301–316.
- (9) Pickering, J. L.; Goodbred, S. L.; Reitz, M. D.; Hartzog, T.R.; Mondal, D. R.; Hossain, Md. S. Late Quaternary Sedimentary Record and Holocene Channel Avulsions of the Jamuna and Old Brahmaputra River Valleys in the Upper Bengal Delta Plain, *Geomorph.* 2014, 227, 123–136. doi:10.1016/j.geomorph.2013.09.021.
- (10) Lambeck, K.; Esat T. M.; Potter, E.-K. Links between climate and sea levels for the past three million years. *Nature*, 2002, 419, 199–206.
- (11) Wiedicke, M.; Kudrass, H.-R.; Hübscher, C. H. Oolitic beach barriers of the last Glacial sea-level lowstand at the outer Bengal shelf. *Marine Geology*, 1999, 157, 7–18.
- (12) McArthur J.M.; Ravenscroft P.; Banerjee D.M.; Milsom J.; Hudson-Edwards K. A.; Sengupta S.; Bristow C.; Sarkar S.; Tonkin S.; Purohit R. How paleosols influence groundwater flow and arsenic pollution: A model from the Bengal Basin and its worldwide implication. *Water Resour. Res.*, 2008, 44: W11411.
- (13) Hoque M.A.; McArthur J.M.; Sikdar P.K. The palaeosol model of arsenic pollution of groundwater tested along a 32 km traverse across West Bengal, India. *Sci. Total Environ.*, 2012, 431: 157–165.
- (14) Hoque MA, McArthur JM and Sikdar PK. 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. *Hydrogeology Journal*, 2014, DOI 10.1007/s10040-014-1139-8.
- (15) Ghosal U., Sikdar P.K. and McArthur J.M.. Palaeosol control of arsenic pollution: the Bengal Basin in West Bengal, India. *Groundwater*, 2015, 53, 588–599. DOI: 10.1111/gwat.12251.
- (16) Sarkar A.; Sengupta, S; McArthur, J. M.; Ravenscroft, P.; Bera, M. K.; Bhushan, R.; Samanta A.; and Agrawal S. Evolution of Ganges–Brahmaputra western delta plain: Clues from sedimentology and carbon isotopes. *Quaternary Science Reviews*, 2009, 28, 2564–2581.
- (17) DPHE 2001. Arsenic contamination of groundwater in Bangladesh. Kinniburgh, D.G. and Smedley, P.L. (eds). Department of Public Health Engineering (DPHE) of Government of Bangladesh and British Geological Survey (BGS) Keyworth.
- (18) McArthur, J. M.; Banerjee, D. M.; Hudson-Edwards, K,A.; Mishra, R.; Purohit, R.; Ravenscroft, P.; Cronin, A.; Howarth, R. J.; Chatterjee, A.; Talukder, T.; Lowry, D.; Houghton, S.; Chadha, D.

- K. Natural organic matter in sedimentary basins and its relation arsenic in anoxic ground water: the example of West Bengal and its worldwide implications. *Appl. Geochem.* 2004, 19,1255–1293.
- (19) McArthur, J. M.; Sikdar, P. K.; Nath, B.; Grassineau, N.; Marshall, J. D.; Banerjee D. M. Sedimentological control on Mn, and other trace elements, in groundwater of the Bengal Delta. *Environ. Sci. Technol.* 2012, 46, 669–676.
- (20) van Geen, A.; Cheng, Z.; Jia, Q.; Seddique, A. A.; Rahman, M. W.; Rahman, M. M.; Ahmed, K. Z. Monitoring 51 community wells in Araihazar, Bangladesh, for up to 5 years: implications for arsenic mitigation. *J. Environ. Sci. Health Part A* 2007, 42, 1729–1740.
- (21) von Brömssen M.; Larsson S. H.; Bhattacharya P.; Hasan M. A.; Ahmed K. M.; Jakariya M.; Sikder M. M. A; Sracek O.; Bivén A.; Doušová B.; Patriarca C.; Thunvik R.; Jacks G. Geochemical characterisation of shallow aquifer sediments of Matlab Upazila, Southeastern Bangladesh Implications for targeting low-As aquifers. *Journal of Contaminant Hydrology*, 2008, 99, 137–149.
- (22) Mukherjee, A.; Fryar, A. E.; Scanlon, B. R.; Bhattacharya, P.; Bhattacharya, A. Elevated arsenic in deeper groundwater of the western Bengal Basin, India: extent and controls from regional to local scale. *Appl. Geochem.* 2011, 26, 600–613.
- (23) Zheng, Y.; Stute, M.; van Geen, A.; Gavrieli, I.; Dhar, R.; Simpson, H. J.; Schlosser, P.; Ahmed, K. M. Redox control of arsenic mobilization in Bangladesh groundwater. *Appl. Geochem.* 2004, 19, 201–214.
- (24) Stollenwerk, K. G.; Breit, G. N.; Welch, A. H.; Yount, J. C.; Whitney, J. W.; Foster, A. L.; Uddin, M. N.; Majumder, R.K.; Ahmed, N. Arsenic attenuation by oxidized aquifer sediments in Bangladesh. *Sci. Tot. Environ.* 2007, 379, 133–150.
- (25) Radloff, K. A.; Zheng, Y.; Michael, H. A.; Stute, M.; Bostick, B. C.; Mihajlov, I.; Bounds, M.; Huq M. R.; Choudhury I.; Rahman M.W.; Schlosser P.; Ahmed K. M.; van Geen A. Arsenic migration to deep groundwater in Bangladesh influenced by adsorption and water demand. *Nat. Geosci.* 2011, 4,793–8.
- (26) McArthur J. M.; Sikdar P. K.; Hoque M. A.; Ghosal U. Waste-water impacts on groundwater: Cl/Br ratios and implications for arsenic pollution of groundwater in the Bengal Basin. *Sci. Total Environ.* 2012, 437, 390–402.

Table S1. Locations, depths, and composition of well waters.

Depths with an asterix were measured after removal of the pump.

Sample	Long	Lat	Depth	Туре		δ18Ο	δD	CI	SO ₄	As	Br
No	WGS84	WGS84	mbgl	of Well	Area	‰	‰	ppm	ppm	ppb	ppb
Piezo at SP											
WB11DS09 65	88.605	22.959	23	ordinary	Nonaghata	-4.7	-32.6	130.0	0.04	92	186
WB11DS12 SP1	88.605	22.959	46	Piezometer	Nonaghata	-5.0	-31.4	6.8	0.01	233	26
WB11DS12 SP2	88.605	22.959	61	Piezometer	Nonaghata	-4.9	-30.9	37.9	0.02	418	51
WB11DS12 SP3	88.605	22.959	91	Piezometer	Nonaghata	-4.8	-31.4	2.7	0.00	2	21
WB11DS12 SP4	88.605	22.959	122	Piezometer	Nonaghata	-4.8	-31.3	3.4	0.01	67	22
Piezo at MP											
WB11DS12 MP1	88.603	22.959	29	Piezometer	Nonaghata	-1.4	-15.0	42.4	0.01	466	61
WB11DS12 MP2	88.603	22.959	46	Piezometer	Nonaghata	-4.9	-29.0	2.5	0.03	4	19
WB11DS12 MP3	88.603	22.959	61	Piezometer	Nonaghata	-4.8	-30.8	2.5	0.02	8	17
WB11DS12 MP4	88.603	22.959	91	Piezometer	Nonaghata	-4.9	-31.6	2.1	0.00	4	16
WB11DS08 61	88.604	22.959	122	Mark II	Nonaghata	-4.4	-30.0	3.7	0.00	59	20
WB12DS07 62A	88.603	22.959	150	Swajaldhara	Nonaghata	-4.5	-29.8	4.0	0.00	25	22
Pleistocene Aquife	-										
UG13 PN 3	88.586	22.994	70	HTW	Panpur			2.1	0.14	0	24
FTP 6	88.583	22.941	76	HTW	Majherpara, Fatehpur			2.6	0.00	0	17
UG12 FP 91	88.592	22.944	85	HTW	Fatehpur			2.4	0.01	0	14
UG13 MUR 9	88.547	22.972	85	HTW	Muragacha	-4.2	-27.2	3.3	0.00	1	19
WB12 DS06 181	88.589	22.950	91	HTW	Ghulam ali para_dasdia	-5.2	-33.2	2.0	0.05	0	12
NG 2	88.567	23.032	95	DIW				5.4	0.01	2	28
UG13 BN 8	88.583	22.924	98	HTW	Bansbana	-4.7	-30.0	3.5	0.00	1	19
WB12 DS06 201	88.597	22.961	110	HTW	Daspara_Dasdia	-5.3	-33.9	1.4	0.06	0	12
UG13 SIM 2	88.520	23.024	110	Mark II	Simurali			9.3	17.82	1	37
Mura 1	88.537	22.973	113		Muragacha			11.3	0.05	1	110
BIS 2	88.568	23.011	115	Mark II	Bizra	-4.0	-26.6	4.9	0.00	5	36
UG13 BA 2	88.533	22.969	116	HTW	Balindi	-3.5	-23.4	3.9	0.03	1	24
WB11DS12 92	88.596	22.958	116	HTW	Daspara	-4.9	-33.8	1.3	0.00	1	12
UG13 MBK 2	88.554	22.946	116	HTW	Mobarekpur	-4.5	-28.1	4.2	0.00	2	40
KRB 1	88.584	22.966	122		Kurambelia			2.9	0.00	0	25
WB11DS08 48A	88.594	22.952	122	HTW	Ghulam Ali para	-4.2	-26.1	22.5	0.01	1	85
UG13 PN 14	88.595	22.989	122	HTW	Panpur	-4.2	-30.5	3.4	0.04	1	38
UG13 GTR 1	88.583	23.020	122	HTW	Gotera	-3.9	-27.2	11.0	0.05	1	68
WB11DS08 8	88.604	22.958	122	HTW	Nonaghata	-3.8	-24.3	385	0.05	2	1361
UG13 MBK 1	88.546	22.947	122	HTW	Jaguli	-4.2	-26.6	6.7	0.00	3	51
UG13 BA 1	88.533	22.969	122	HTW	Balindi			5.1	0.00	4	37
RTN 2	88.566	22.895	122	HPW	Ruitanpur	-4.6	-30.6	2.6	0.00	2	22

SMH 2	88.549	22.954	122	HPW	Simhat Primary School	-4.0	-26.6	5.0	0.00	2	43
PNP	88.579	22.996	128	DIW	Panpur	-4.5	-30.1	4.0	0.02	1	35
FTP 2	88.592	22.929	130	Mark II	Dakshin para, Fatehpur			7.0	0.00	5	34
KRB 10	88.577	22.983	130	Mark II	Nischintapur, Kurambelia			2.9	0.00	3	15
UG13 PN 16	88.591	22.984	133	HTW	Panpur	-5.3	-34.6	2.8	0.23	0	17
UG13 PN 17	88.593	22.984	134	HTW	Panpur	-4.7	-30.4	6.0	0.00	1	41
UG13 BA 3	88.534	22.970	134	HTW	Balindi			4.7	0.00	5	37
Molla 3	88.585	22.950	134		Mollabelia			1.6	0.00	7.7	13
MBL	88.590	22.986	134	DIW	Mullebelia	-4.8	-32.9	2.6	0.20	1	23
NRP 2	88.521	23.024	137	DIW		-4.5	-29.9	4.7	0.69	1	38
NRP 1	88.533	23.018	140	DIW		-4.5	-30.0	6.4	0.09	1	36
JRP	88.533	23.024	146	DIW		-5.0	-32.2	2.7	0.67	2	37
WB12 DS06 184	88.589	22.950	152	Mark II		-5.3	-33.3	1.4	0.06	1	14
UG13 PN 44	88.585	22.989	152	Mark II	Panpur	-5.0	-32.0	3.3	0.00	1	17
UG13 PN 15	88.589	22.987	152	DIW	Panpur	-5.1	-33.1	2.5	0.11	1	22
UG13 SIM 1	88.521	23.025	152	Mark II	Simurali			5.3	0.05	1	31
UG13 PN 53	88.577	22.998	152	Mark II	Panpur	-5.1	-31.5	4.2	0.00	1	20
WB12 DS06 182	88.589	22.950	152	Mark II		-5.2	-32.7	2.4	0.05	2	16
UG 13 HKN 11	88.529	22.942	152	Mark II	Hatikanda	-4.7	-30.7	6.9	0.03	5	28
UG13 PN 52	88.581	22.993	152	HTW	Panpur	-4.5	-30.9	3.5	0.00	7	106
UG13 BA 10	88.533	22.968	152	Mk II	Balindi	-4.2	-27.7	4.7	0.03	8	42
UG12 FP 29	88.584	22.942	156	Mark II	Fatehpur			2.8	0.00	2	13
DIG 2	88.540	22.944	165	Mark II	Digha, Harighata	-4.5	-29.0	9.7	0.57	1	78
ANN	88.544	22.927	165	Mark II	Harighata	-3.5	-20.9	69.6	14.04	1	277
FAT 1	88.583	22.945	165	Mark II	Harighata	-3.6	-22.0	34.6	4.92	3	124
FAT 2	88.584	22.945	165	Mark II	Harighata	-3.6	-22.0	44.9	3.99	3	173
DIG 1	88.543	22.943	165	Mark II	Dhiga village	-4.3	-27.9	5.4	0.01	1	40
UG13 PN 18	88.594	22.983	168	Mark II	Panpur	-4.9	-30.8	8.0	0.00	0	37
UG12 FP 72	88.592	22.929	171	Mark II	Fatehpur			7.0	0.00	5	30
CHP	88.508	23.026	171	DIW		-4.9	-31.8	3.1	1.38	2	35
NG 1	88.559	23.034	177	DIW		-4.5	-29.9	6.2	0.09	1	35
RMP	88.510	23.020	183	DIW		-4.8	-32.0	2.6	0.68	2	36
UG12 FP 10	88.585	22.944	195	Mark II	Fatehpur			32.5	5.31	1	113
BIS 1	88.573	22.996	274	DIW	Bizra	-4.8	-31.7	2.6	0.00	1	22

Pleistocene Aquifers at 35 - 70 mbgl under a Palaeo-interfluve

Others lacking stable isotope data can be found in Ghosal et al. 2015												
UG13 MBK 10	88.549	22.945	43	HTW	Mobarekpur	-4.0	-27.7	14.6	1.65	2	92	
UG13 MBK 12	88.550	22.944	43	HTW	Mobarekpur	-2.1	-16.6	5.1	0.02	1	45	
WB12 DS06 208	88.603	22.958	46	HTW	Kuthipara_Dasdia	-5.1	-31.4	2.3	0.05	0	18	
WB11DS08 68	88.604	22.960	46	HTW	Nonaghata	-5.0	-31.1	2.4	0.00	1	16	
UG13 PN 45	88.583	22.992	46	HTW	Panpur	-4.8	-30.2	2.6	0.00	0	36	
UG13 PN 33	88.586	22.987	46	HTW	Panpur	-4.5	-27.6	2.1	0.00	0	16	
UG13 MBK 11	88.550	22.944	46	HTW	Mobarekpur	-2.2	-17.2	6.7	0.50	1	32	
SMH 1	88.546	22.954	46	HPW	Simhat	-4.88	31.75	12.3	1.60	0	320	
BIS 4	88.575	23.000	46	SIW	Bizra	-4.50	30.63	2.5	0.09	1	29	
WB12 DS06 207	88.603	22.958	49	HTW	Nonaghata	-5.0	-31.5	2.5	0.10	1	18	

WB12 DS06 206	88.603	22.957	49	HTW	Nonaghata	-4.8	-30.0	3.2	0.10	0	16
UG13 PN 42	88.589	22.993	49	SIW	Panpur	-4.5	-29.4	3.1	0.01	2	36
UG13 PN 20	88.596	22.991	49	HTW	Panpur	-4.2	-27.1	4.4	0.00	5	29
UG13 PN 23	88.595	22.991	49	HTW	Panpur	-4.2	-27.8	4.7	0.00	0	36
UG13 PN 24	88.595	22.990	49	HTW	Panpur	-4.2	-28.1	4.3	0.01	1	39
UG13 PN 22	88.596	22.991	49	HTW	Panpur	-4.1	-27.3	4.1	0.00	0	46
UG13 PN 21	88.595	22.991	49	HTW	Panpur	-3.9	-27.0	4.1	0.00	0	39
UG13 PN 27	88.595	22.989	50	HTW	Panpur	-4.4	-29.6	2.8	0.04	1	50
UG13 PN 50	88.584	22.991	52	HTW	Panpur	-4.7	-28.9	3.3	0.02	0	27
UG13 PN 25	88.595	22.990	52	HTW	Panpur	-4.3	-28.6	5.1	0.00	0	37
WB11DS12 60	88.597	22.956	55	SIW	Daspara	-5.1	-33.8	1.8	0.33	0	11
UG13 PN 31	88.594	22.988	55	SIW	Panpur	-4.4	-29.2	2.1	0.01	0	37
UG13 PN 28	88.595	22.989	55	HTW	Panpur	-4.4	-29.9	3.0	0.00	0	66
UG13 PN 43	88.589	22.995	55	SIW	Panpur	-3.8	-26.2	4.3	0.00	7	31
UG13 PN 26	88.595	22.990	56	HTW	Panpur	-4.0	-28.4	3.8	0.00	0	61
WB11DS12 165	88.592	22.953	58	SIW	Nonaghata	-5.0	-33.4	1.9	0.02	1	10
UG13 PN 47	88.583	22.991	61	HTW	Panpur	-4.7	-30.6	2.5	0.00	1	29
UG13 PN 46	88.583	22.991	61	HTW	Panpur	-4.7	-29.5	3.2	0.01	1	26
UG13 PN 48	88.583	22.991	61	HTW	Panpur	-4.4	-27.1	3.1	0.00	0	21
SAT	88.581	23.012	61	SIW	Satberia, in large paddy.	-3.67	25.39	6.2	0.00	1	46

Pleistocene Aquifers at > 70 mbgl under a Palaeo-interfluvial Margin												
WB11DS09 07	88.604	22.958	70	HTW	Nonaghat	a		8.3	0.00	56	31	
UG13 PN 13	88.594	22.988	73	*HTW	Panpu	ır -4.5	-29.8	2.5	0.03	13	30	
UG13 MBK 8	88.549	22.945	82	*HTW	Mobarekpu	ır -3.1	-21.4	6.6	0.04	19	34	
UG12 FP 182	88.586	22.941	91	HTW	Fatehpu	ır		4.2	0.01	1	16	
UG13 MBK 6	88.549	22.944	91	HTW	Mobarekpu	ır -3.6	-24.8	6.2	0.00	17	31	
UG12 FP 92	88.592	22.944	94	*HTW	Fatehpu	ır		6.1	0.00	115	19	
UG12 FP 169	88.588	22.939	97	*Mark II	Fatehpu	ır		4.2	0.00	36	15	
UG13 MBK 3	88.549	22.944	98	*HTW	Mobarekpu	ır -2.9	-20.9	6.8	0.00	15	32	
UG12 FP 100	88.591	22.945	107	*Mark II	Fatehpu	ır		7.1	0.01	34	26	
BNB 4	88.583	22.931	110		Bansban	a		10.4	0.00	42	36	
UG 13 HKN 9	88.517	22.937	116	HTW	Hatikand	a -5.5	-35.9	16.0	0.19	2	33	
UG13 BN 14	88.584	22.925	118	*Mark II	Bansban	a -4.6	-30.1	11.2	0.00	135	29	
UG13 BHA 9	88.548	22.927	119	*Mark II	Bhabanipu	ır -4.3	-27.8	8.2	0.01	1	23	
Fateh 3	88.592	22.945	122		Fatehpu	ır		2.2	0.00	27	22	
UG13 SBN 17	88.577	22.946	125	Mark II	Subarnapu	ır -4.9	-32.0	3.9	0.00	54	22	
UG12 FP 30	88.584	22.941	126	*HTW	Fatehpu	ır		3.8	0.00	108	17	
UG13 SBN 15	88.573	22.956	128	*Mark II	Subarnapu	ır -4.7	-30.3	23.1	0.00	82	41	
KRB 4	88.595	22.972	134		Kurambeli	a		3.7	0.00	24	29	
UG13 PN 12	88.577	22.986	136	*HTW	Panpu	ır		23.1	0.02	25	76	
UG13 SBN 16	88.580	22.956	144	*HTW	Subarnapu	ır -4.4	-28.6	8.9	0.00	37	42	
UG13 KM 1	88.594	22.974	155	*Mark II	Kurambeli	a		4.5	0.00	30	30	
UG13 MBK 7	88.549	22.944	156	*HTW	Mobarekpu	ır		5.5	0.01	25	31	

Pleistocene Aquife	ers 35 - 70 m	nbgl under a l	Palaeo-interfl	uvial Margin							
Others lacking sta	ble isotope	data can be f	ound in Ghos	sal et al. 2015							
UG13 MB 28	88.575	22.963	40	HTW	Mollabelia (North)			3.9	0.22	2	12
UG13 MUR 3	88.548	22.972	40	HTW	Muragacha	-3.4	-25.1	14.4	1.14	4	24
WB11DS12 143	88.604	22.958	43	HTW	Nonaghata			6.2	0.00	0	21
Molla 2	88.582	22.952	43		Mollabelia			6.6	1.10	1	20
UG13 BN 3	88.583	22.925	43	HTW	Bansbara			6.1	0.00	1	24
UG12 BB 55	88.598	22.963	43	HTW	Bamunberia			44.5	12.36	15	40
UG13 UR 4	88.572	22.969	46	HTW	Uttar Rajapur			1.9	0.07	1	8
UG13 BJ 2	88.572	22.998	47	*HTW	Bijra_Panchanantala			2.0	0.00	19	12
WB11DS12 150	88.599	22.950	48	HTW	Nonaghata			3.4	0.00	18	18
WB11DS12 168	88.602	22.953	49	SIW	Nonaghata	-3.6	-25.8	9.2	0.43	1	27
UG13 UR 2	88.574	22.979	49	HTW	Uttar Rajapur			3.5	3.36	1	18
UG13 UR 1	88.574	22.979	49	HTW	Uttar Rajapur			4.6	4.90	1	22
UG13 BJ 1	88.572	22.998	49	HTW	Bijra_Panchanantala			3.5	0.33	3	12
UG13 PN 10	88.577	22.986	49	HTW	Panpur			15.1	0.01	0	33
WB11DS12 136	88.604	22.957	50	HTW	Nonaghata			4.5	0.02	0	17
UG13 MUR 2	88.548	22.972	50	HTW	Muragacha	-3.6	-24.8	22.1	5.26	1	34
UG12 FP 178	88.587	22.940	50	HTW	Fatehpur			3.4	0.03	1	15
WB12 DS06 205	88.603	22.957	52	HTW	Nonaghata	-4.5	-27.5	2.9	0.01	0	11
UG13 MUR 5	88.549	22.972	52	HTW	Muragacha	-4.2	-29.0	6.4	0.00	4	16
UG12 FP 98	88.592	22.945	54	*HTW	Fatehpur			2.1	0.00	13	18
UG13 MB 4	88.582	22.950	55	*HTW	Mollabelia (South)			2.0	0.00	15	13
UG13 PN 30	88.595	22.988	55	HTW	Panpur	-4.4	-28.9	4.4	1.37	0	57
UG13 UR 3	88.573	22.978	55	HTW	Uttar Rajapur			9.9	3.60	0	20
UG12 FP 1	88.584	22.946	55	HTW	Fatehpur			3.3	0.00	1	11
UG13 MUR 1	88.548	22.972	55	HTW	Muragacha	-3.8	-25.2	23.6	1.07	2	39
UG13 MB 3	88.581	22.949	55	HTW	Mollabelia (South)			3.4	0.01	6	12
UG13 MB 1	88.581	22.949	55	HTW	Mollabelia (South)			2.6	0.00	10	11
UG13 MB 26	88.576	22.963	58	HTW	Mollabelia (North)			9.1	0.61	1	19
UG13 MB 25	88.576	22.963	58	HTW	Mollabelia (North)			5.4	0.59	1	16
UG13 BN 1	88.583	22.924	61	*SIW	Bansbara			4.2	0.02	14	18
UG12 FP 4	88.585	22.946	67	HTW	Fatehpur			2.1	0.00	0	8
Pleistocene Aquife	ers at > 70 m	n depth under	Deep Palaed	o-channels							
UG13 AB 2	88.571	22.938	79	HTW	Atbeharia	-4.2	-29.3	20.2	3.95	8	26
UG13 BN 7	88.583	22.924	98	*HTW	Bansbana	-4.5	-29.3	7.2	0.00	133	22
UG13 AB 10	88.570	22.941	101	*Mark II	Atbeharia	-4.7	-30.2	14.2	3.36	72	27
UG13 SBN 10	88.566	22.948	104	*Mark II	Subarnapur	-4.6	-29.2	14.6	0.00	60	37
UG13 UR 14	88.564	22.981	105	*Mark II	Uttar Rajapur	-5.1	-33.1	27.9	0.38	38	51
DOG	88.581	22.883	107	Mark II	Dogachia	-4.47	29.72	3.2	0.00	22	19
BJR 3	88.555	23.013	110		Bijra			5.9	0.00	16	35
UG13 SBN 13	88.567	22.945	113	HTW	Subarnapur	-4.3	-27.8	5.7	0.00	52	13

4.4

4.7

6.0

3.1

-32.3

-31.7

-32.1

-4.9

-4.9

-4.7

0.31

0.02

0.05

0.00

Sonakhali

Kuthipara

Subarnapur

Sri krishnapur

17

23

29

14

10

13

16

29

UG13 SON 12

WB11DS08 21

UG13 SBN 2

UG13 SKP 1

23.002

22.953

22.951

22.911

88.554

88.609

88.564

88.567

122

122

122

122

Mark II

HTW

 HTW

HTW

UG13 SB 10	88.636	22.960	122	HTW	Subuddipur			15.9	0.00	33	32
UG13 GTR 18	88.602	22.999	122	HTW	Dakshin Panchpota			4.0	0.00	102	18
BM 2	88.559	22.925	122	STW	Brahmanpur	-4.12	28.97	2.5	0.01	36	23
PRB 1	88.545	22.898	122	HPW	Netai Ghosh, village ?	-4.06	28.20	4.3	0.00	36	29
UG12 JK 82	88.613	22.962	122	HTW	Julkarampur			24.3	0.00	41	50
UG12 JK 231	88.618	22.957	122	HTW	Julkarampur			17.5	0.00	64	39
UG12 BB 81	88.600	22.968	122	HTW	Bamunberia			3.9	0.00	100	29
UG12 BB 11	88.599	22.962	126	*HTW	Bamunberia			2.7	0.00	59	14
ВНК	88.591	22.873	128	Mark II	Bhurkundra	-3.64	23.54	5.6	0.01	33	24
MSD 1	88.587	23.013	134	HPW	Mohishdang	-4.15	29.15	2.4	0.01	103	22
WB12 DS06 216	88.609	22.954	134		Kuthipara	-4.9	-31.5	22.0	0.03	1	82
UG 13 SON 9	88.557	23.016	134	HTW	Sonakhali/ Baksha	-4.5	-29.9	6.7	0.00	40	32
UG13 GTR 22	88.584	23.025	134	HTW	Ghetugachi			3.5	0.05	59	13
WB11DS12 14	88.604	22.960	134	HTW	Nonaghata	-4.4	-30.6	16.1	0.03	61	65
UG13 SBN 12	88.567	22.945	134	HTW	Subarnapur	-5.2	-34.0	4.3	0.00	120	20
UG13 GTR 17	88.603	23.000	134	HTW	Dakshin Panchpota			4.0	0.00	134	17
UG13 GTR 16	88.601	23.002	134	HTW	Dakshin Panchpota			4.1	0.00	147	17
UG13 SBN 11	88.565	22.947	134	HTW	Subarnapur	-3.6	-25.5	34.9	1.51	157	48
UG13 JB 1	88.635	22.975	134	HTW	Jalkarbhomra	-4.8	-30.2	12.8	0.00	184	48
WB13 SK 37	88.644	22.954	137	HTW	Mahadebpur			13.7	0.01	90	29
2014 SON 11b	88.554	23.002	140	HPW	Sonarkali	-4.4	29.59	4.3	2.04	9	23
UG13 SBN 6	88.564	22.950	143	HTW	Subarnapur	-4.9	-31.2	4.8	0.00	51	31
UG13 MD1	88.609	22.913	146	Mark II	Mali Danga			3.0	0.00	57	15
UG 13 SON 1	88.558	23.015	152	Mark II	Sonakhali/ Baksha	-4.23	-28.7	6.0	0.01	30	29
UG 13 SON 10	88.555	23.013	152	Mark II	Sonakhali/ Baksha	-4.56	-30.2	4.5	0.00	13	24
UG13 BIR 11	88.542	22.995	152	Mark II	Birohi	4.00	00.2	2.8	0.03	65	17
UG13 DB 2	88.658	22.945	152	Mark II	Dasberia			1.5	0.02	51	9
UG13 DKD 2	88.606	22.938	152	Mark II	Dakshin dattapara			3.0	1.25	3	21
UG13 DKD 9	88.600	22.944	152	Mark II	Dakshin dattapara			4.8	0.00	26	21
UG13 GN 22		22.959	152		·			212.4	0.00	95	762
	88.678			Mark II	Gopinathpur						
UG13 GN 24	88.674	22.957	152	Mark II	Gopinathpur	4.71	21.0	184.9	0.00	93	663
UG13 GTR 11	88.595	23.016	152	Mark II	Ramkestopur	-4.71	-31.0	4.4	0.01	76	18
UG13 GTR 13	88.602	23.005	152	Mark II	Dakshin Panchpota/Kadambagachi	-4.67	-30.2	3.2	0.00	113	17
UG13 GTR 15	88.603	23.003	152	Mark II	Dakshin Panchpota	-4.79	-30.6	3.5	0.00	93	17
UG13 GTR 19	88.599	23.000	152	Mark II	Dakshin Panchpota	-4.52	-30.6	3.4	0.00	125	18
UG13 GTR 20	88.598	23.001	152	Mark II	Dakshin Panchpota	-4.68	-29.9	4.4	0.00	83	20
UG13 GTR 23	88.604	23.011	152	Mark II	Kadambagachi	-4.72	-30.7	6.1	0.00	98	23
UG13 GTR 24	88.601	23.006	152	Mark II	Dakshin Panchpota	-4.80	-30.9	4.4	0.00	28	20
UG13 GTR 4	88.588	23.020	152	Mark II	Ramkestopur(Gotera)	-4.53	-30.2	2.8	0.01	82	16
UG13 JB 10	88.635	22.974	152	Mark II	Jalkarbhomra	-4.56	-30.5	66.5	0.02	65	232
UG13 JB 3	88.634	22.975	152	Mark II	Jalkarbhomra	-4.68	-30.0	68.3	0.00	50	225
UG13 JB 7	88.631	22.977	152	Mark II	Jalkarbhomra	-4.91	-31.9	35.3	0.00	35	122
UG13 PD 10	88.622	22.898	152	Mark II	Payradanga/Parulia	-4.85	-31.5	2.4	0.00	49	14
UG13 PK 12	88.564	22.936	152	Mark II	Panchkaun	-4.83	-30.3	3.8	0.00	326	30
UG13 PN 19	88.599	22.980	152	Mark II	Panpur Haringhata-Jaguli Rd	-4.90	-30.8	6.8	0.00	20	39
UG13 SBN 14	88.561	22.956	152	Mark II	Kaltala/Subarnapur	-4.38	-29.2	12.1	0.01	32	47
UG13 SKP 6	88.568	22.913	152	Mark II	Sri krishnapur	-4.56	-29.2	3.6	0.00	26	15
UG13 GTR 3	88.585	23.021	152	Mark II	Ramkestopur(Gotera)	-4.7	-30.7	4.1	0.00	77	15

AYP	88.546	22.986	165	Mark II	School well, Digha, Harighata	-5.37	-34.3	1.9	0.00	275	16
BDK	88.637	22.908	165	Mark II	School, Harighata	-3.08	21.35	3.2	0.00	122	29
HPK	88.613	22.950	165	Mark II	School, Haripurkuria	-4.74	30.61	12.9	0.03	1	63
HRG	88.568	22.962	165	Mark II	Harighata	-3.68	21.70	21.5	0.17	3	85
NYP	88.558	22.974	165	Mark II	Harighata, Dipankar's well	-4.44	31.62	1.8	0.02	41	12
UG12 JK 283	88.615	22.961	195	HTW	Julkarampur	-3.82	23.29	480.0	0.01	3	1778
DR 1	88.566	22.972	237	Mark II	Harighata	-2.88	18.90	33.8	34.51	13	125
SL 1	88.564	22.962	249	Mark II	Harighata	-3.32	20.39	30.9	17.25	2	118

Holocene Aquifers 35 - 70 mbgl in Deep Palaeo-channels												
Others lacking stal	ble isotope	data can be	found in Ghos	al et al. 2015								
UG13 JB 6	88.633	22.976	37	HTW	Jalkarbhomra	-6.0	-38.6	16.7	3.15	28	15	
UG13 PD 6	88.621	22.898	37	HTW	Payradanga/Parulia	-5.3	-35.6	35.6	15.95	5	43	
UG13 AB 8	88.571	22.938	37	HTW	Atbeharia	-4.9	-33.6	31.1	10.68	3	37	
UG13 GTR 2	88.600	23.000	37	HTW	Panchpota	-4.7	-29.3	5.8	0.00	201	26	
UG13 GTR 10	88.594	23.017	37	HTW	Ramkestopur	-4.4	-27.8	70.5	11.83	62	84	
UG13 SKP 5	88.568	22.913	37	HTW	Sri krishnapur	-3.4	-24.4	23.9	13.14	41	61	
UG13 GTR 8	88.593	23.018	37	HTW	Ramkestopur/Gotera/Kadamgachi	-3.1	-20.8	7.2	0.00	42	27	
UG13 PK 3	88.566	22.934	37	HTW	Panchkaun	-2.6	-17.8	7.8	0.00	93	46	
UG13 BN 13	88.581	22.925	40	*HTW	Bansbana	-4.4	-27.9	17.7	0.00	88	37	
UG13 PK 9	88.566	22.933	43	HTW	Panchkaun	-3.2	-21.9	7.6	0.06	18	34	
UG13 BIR 5	88.541	22.991	46	HTW	Birohi	-5.7	-37.6	11.8	0.01	103	14	
UG13 BIR 2	88.549	22.991	46	HTW	Birohi	-5.7	-37.7	16.4	10.51	53	11	
UG13 BIR 10	88.547	22.991	46	HTW	Birohi	-5.7	-36.9	20.4	1.65	35	20	
UG13 BIR 9	88.546	22.991	46	HTW	Birohi	-5.6	-36.7	19.6	1.50	61	11	
UG 13 SON 7	88.558	23.016	46	HTW	Sonakhali/ Baksha	-5.0	-32.5	33.6	0.00	31	86	
UG13 BHA 10	88.548	22.927	46	HTW	Bhabanipur	-4.8	-30.3	73.4	27.56	6	55	
WB11DS12 169	88.608	22.954	46	SIW	Kuthipara	-3.9	-27.3	5.5	0.65	114	16	
UG13 PK 8	88.566	22.933	46	HTW	Panchkaun	-3.7	-24.8	6.7	0.00	22	34	
UG13 PK 1	88.566	22.933	46	HTW	Panchkaun	-3.4	-23.2	6.0	0.01	40	35	
BM 1	88.559	22.925	46	SIW	Bizra	-4.26	29.12	3.5	0.01	43	24	
WB11DS12 170	88.606	22.953	49	SIW	Kuthipara	-4.2	-27.8	2.5	0.02	92	13	
UG13 MUR 7	88.548	22.973	49	HTW	Muragacha	-3.8	-25.2	33.3	3.44	2	45	
PRB 2	88.545	22.898	49	HPW	Netai Ghosh, ?	-4.28	29.30	12.2	0.00	65	59	
WB11DS08 29	88.610	22.952	52	HTW	Kuthipara	-4.1	-26.1	2.3	0.04	86	12	
UG13 SON 8	88.558	23.016	52	HTW	Sonakhali/ Baksha	0.0	-4.5	20.9	0.00	10	67	
UG13 PD 4	88.621	22.898	53	HTW	Payradanga/Parulia	-4.3	-30.1	19.4	13.76	1	23	
UG13 AB 4	88.570	22.938	55	SIW	Atbeharia	-4.3	-29.1	33.8	0.85	26	49	
UG13 BN 10	88.581	22.924	55	HTW	Bansbana	-4.2	-27.9	40.0	0.00	123	68	
UG13 BN 12	88.581	22.925	58	HTW	Bansbana	-4.3	-27.1	19.8	0.00	257	32	
UG13 GTR 21	88.583	23.022	58	HTW	Gotera	-3.6	-24.2	20.1	5.23	27	37	
UG13 BN 11	88.581	22.925	58	HTW	Bansbana	-3.4	-23.5	43.8	2.85	117	63	
PH 2	88.565	22.929	61	STW	Panchkauni	-3.89	25.61	4.0	0.02	198	36	
UG13 SKP 4	88.568	22.912	61	HTW	Sri krishnapur	-3.1	-24.3	26.9	4.79	28	47	
MSD 2	88.589	23.013	64	SIW	Mohishdang	-3.57	25.53	5.3	0.00	95	32	