Water Resources Research

RESEARCH ARTICLE
10.1029/2017WR022485

Key Points:
- Arsenic levels in shallow wells increase over the 35 km separating the banks of the Brahmaputra River from the Naga foothills in Assam
- The thickness of clay capping the >12-kyr-old aquifer also increases <1 to 60 m from the banks of the river to the foothills
- A model consistent with groundwater heads and ages suggests that arsenic was flushed out of the aquifer more effectively near the river

Supporting Information:
- Supporting Information S1

Correspondence to:
R. Choudhury and A. van Geen,
runti.choudhury@gmail.com;
avangeen@ldeo.columbia.edu

Citation:
Choudhury, R., Nath, B., Khan, M. R., Mahanta, C., Ellis, T., & van Geen, A. (2018). The impact of aquifer flushing on groundwater arsenic across a 35-km transect perpendicular to the upper Brahmaputra River in Assam, India. *Water Resources Research*, *54*, 8160–8173. https://doi.org/10.1029/2017WR022485

Received 27 DEC 2017
Accepted 24 JUL 2018
Accepted article online 3 AUG 2018
Published online 22 OCT 2018

The Impact of Aquifer Flushing on Groundwater Arsenic Across a 35-km Transect Perpendicular to the Upper Brahmaputra River in Assam, India

R. Choudhury[1,2], B. Nath[3], M. R. Khan[4], C. Mahanta[1], T. Ellis[3], and A. van Geen[3]

[1]Department of Civil Engineering, Indian Institute of Technology, Guwahati, India, [2]Now at Department of Geological Sciences, Gauhati University, Gauhati, India, [3]Lamont-Doherty Earth Observatory, Columbia University, Palisades, NY, USA, [4]Department of Geology, University of Dhaka, Dhaka, Bangladesh

Abstract Well testing in the floodplain of the Brahmaputra River in Golaghat and Jorhat districts of Assam, India, shows that groundwater arsenic (As) concentrations increase with distance from the river. To establish the origin of this pattern, an additional 900 wells <60-m deep were tested for As and nine sites were drilled along a 35-km transect perpendicular to the river. The field data show no relation between groundwater As concentrations ranging from <1 to 660 μg/L along the transect and (a) As concentrations of <1–5 mg/kg in cuttings of aquifer sand recovered while drilling or (b) the degree of reduction of iron oxides in these cuttings. The drilling indicates, however, a marked increase in the thickness of a clay layer capping the aquifer starting from <1–5 m near the river to over 60 m at the most distant site toward the base of the Naga foothills. Organic radiocarbon ages of 18–46 kyr obtained from all but one of 13 clay samples indicate pre-Holocene deposition of the underlying sands across the entire transect. Radiocarbon ages of dissolved inorganic carbon of 0.2, 4.7, and 17.8 kyr were measured in groundwater from three monitoring wells installed to 30–60-m depth at distances of 10, 20, and 40 km from the river, respectively. A conceptual groundwater flow model consistent with monitored heads and groundwater ages suggests that thick clay layers capping the aquifer further from the river inhibited flushing of the aquifer and, as a result, preserved higher As levels in groundwater.

Plain Language Summary A large fraction of the rural population of South Asia living in river floodplain areas is exposed to toxic levels of arsenic by drinking well water. This study combines field data from the Indian state of Assam with a groundwater flow model to understand the mechanisms that control the distribution of arsenic in groundwater. A field kit was used to document groundwater arsenic concentrations that increase from the banks of the Brahmaputra River to the foothills of nearby mountains. Drilling showed that the sandy aquifer becomes isolated from replenishment with local surface water along the same transect by an increasingly thick impermeable capping clay layer. A simple groundwater flow model of the area tuned to match available data indicates that aquifers near the Brahmaputra River were flushed of much of their initial arsenic content by recharge with low-arsenic surface water over thousands of years. With increasing distance from the river, this flushing was progressively inhibited by a layer of clay layer capping the aquifer and elevated arsenic remained high as a result. Mitigation efforts to reduce arsenic exposure should take into account that the distribution of arsenic in well water evolves slowly and is therefore unlikely to change very soon.

1. Introduction

Elevated arsenic (As) concentrations in well water are a major public health concern for over 100 million people across South and Southeast Asia (Ravenscroft et al., 2009; Smedley & Kinniburgh, 2002). Reductive dissolution of Fe (III) oxyhydroxide minerals has emerged as the leading mechanism for the release of As to groundwater (Berg et al., 2001; Bhattacharya et al., 1997; Fendorf et al., 2010; McArthur et al., 2004; Nickson et al., 1998; Postma et al., 2012). Yet several key factors including the relative importance of sedimentary and advected sources of reduced carbon for the dissolution of Fe (III) oxides remain poorly understood (Datta et al., 2011; Meharg et al., 2008; Neumann et al., 2010). Some studies provide evidence of a very shallow source of reactive carbon and/or reactive Fe oxides, both of which are required to release sequestered As from aquifer sediments to groundwater (Neumann et al., 2010; Polizzotto et al., 2008; Stuckey et al., 2016). Other observations suggest that advected organic matter and exchange of As with the aquifer sediment

continue to play an important role further along the groundwater flow path (Mailloux et al., 2013; Postma et al., 2012; Radloff et al., 2017). This is probably one of the reasons that the documented heterogeneity of As concentrations in groundwater across a wide range of spatial scales remains poorly understood (Ahmed et al., 2004; BGS/DPHE, 2001; McArthur et al., 2008; Winkel et al., 2011).

In various attempts to understand the origin of heterogeneity, some have linked groundwater As occurrences to paleogeomorphology, near-surface geology, depositional history, and hydrogeology (Donselaar et al., 2017; Hoque et al., 2009; McArthur et al., 2011; Nath et al., 2005; van Geen et al., 2008). Weinman et al. (2008) pointed out that floodplain evolution over the past few thousand years seemed to control groundwater As heterogeneity in shallow aquifers of a study area in Bangladesh. Aziz et al. (2008) proposed for the same area that the spatially variable permeability of near-surface sediments contributed to aquifer As heterogeneity by influencing recharge rates of the underlying aquifer. Elsewhere in the Bengal Basin, Nath et al. (2010) also showed that thick clay capping high-As aquifers inhibited direct recharge. Such observations led van Geen et al. (2008) to propose that recharge and transport over time reduce the concentration of exchangeable As in Holocene ($<$12-kyr-old) sediment and therefore also the concentration of As in groundwater in equilibrium with the solid phase.

Invoking processes acting over longer time scales, BGS/DPHE (2001) and McArthur et al. (2008, 2011) attributed lateral heterogeneity to elevated groundwater As levels in Holocene aquifers deposited within paleochannels formed by erosion during sea level low stands. In contrast, low-As concentrations are characteristic of Pleistocene ($>$12-kyr) aquifers preserved within paleointerfluves. Low-As concentrations in these Pleistocene aquifers have been attributed to reoxidation of Fe oxides and/or flushing of an exchangeable As fraction during low sea level stands (BGS/DPHE, 2001; Zheng et al., 2005).

Elevated levels of As in groundwater of the upper Brahmaputra floodplains were first reported more recently than elsewhere in South Asia (Borah et al., 2009; Chetia et al., 2011; Choudhury et al., 2015; Mahanta et al., 2015; Singh, 2004). Singh (2004) first suggested that abundant clay layers with a high organic carbon content in some parts of the state might have led to the release of As to groundwater. In the affected district of Jorhat (Figure 1), on the southern bank of the Brahmaputra River, Chakraborti et al. (2004) and Goswami et al. (2013) reported dissolved As concentrations of almost 500 μg/L. Verma et al. (2016) pointed out the greater abundance of clay at a drill site in Jorhat compared to two drill sites on the northern bank of the Brahmaputra but tentatively attributed higher As concentration in Jorhat to differences in tectonic history and sediment provenance. A joint study by United Nations International Children's Emergency Fund (UNICEF) and the Assam Public Health Engineering Department (APHED) conducted with field kits documented As concentration above the World Health Organization guideline of 10 μg/L in 76 administrative blocks (Mahanta et al., 2015). Out of a total of 56,180 tested, 29% of wells contained As $>$10 μg/L and 8% wells contained As $>$50 μg/L (Figure 1a). In 2012, the national Bureau of Indian Standards adopted a two-standard policy for As in drinking water: 10 μg/L as a recommended acceptable limit and 50 μg/L as a permissible limit in the absence of alternative sources that contain less As (Bureau of Indian Standards, 2012). The Assam PHED refers to these standards in its efforts to provide safe drinking water to rural communities (Assam Public Health Engineering Department, 2017).

The main motivation for selecting the Jorhat area south of the Brahmaputra River for further study was that As concentrations increase with distance from the river instead of decreasing with distance from the river, as documented further downstream in the Bengal Basin (BGS/DPHE, 2001; van Geen et al., 2008). In this paper, we first document in more detail the regional distribution of As in aquifers $<$60-m deep in a region straddling the border between Golaghat and Jorhat district with over 900 well tests using a field kit (Figure 1b), a subset of which were verified by laboratory analysis. We then present lithological and geochemical data collected while manually drilling nine sites along the 35-km transect that extends from the banks of the Brahmaputra River to the foothills of the Naga Patkai Hill ranges. The field data are complemented by radiocarbon dating of organic carbon in a set of clay cuttings recovered along the transect and of dissolved inorganic carbon in groundwater samples collected from monitoring wells installed along the same transect. The various observations are then linked with a simplified 2-D cross-sectional groundwater flow model that extends from the center of the Brahmaputra River to the foothills of the Naga Patkai Hill ranges. The model shows that groundwater As concentrations increasing with distance from the river can be explained by differences in surface permeability and recharge rates. This leads us to propose that the new observations

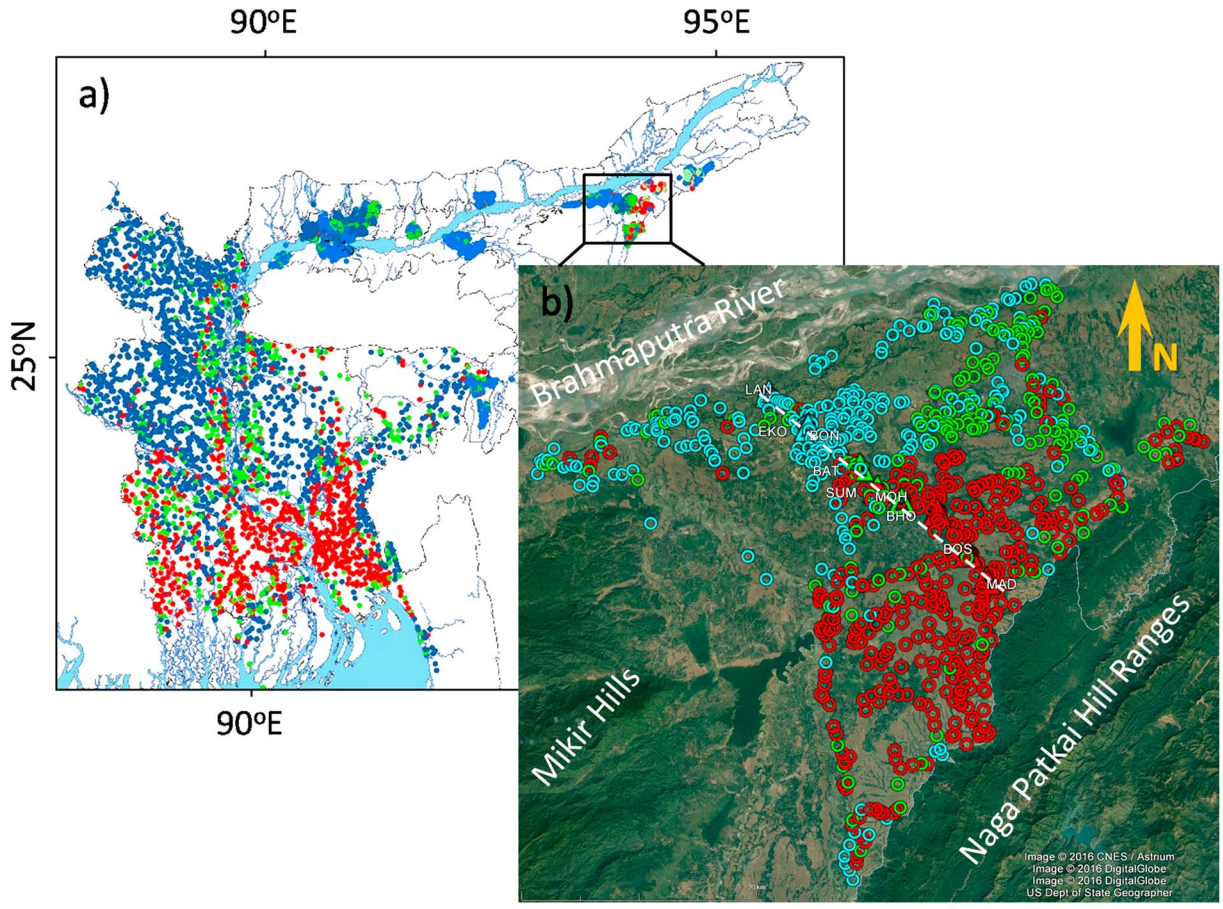

Figure 1. (a) Map showing As concentrations in shallow wells (<60-m depth) in the upper Brahmaputra River basin in Assam (Mahanta et al., 2015) along with the As distribution in the lower Brahmaputra River basin in Bangladesh (BGS/DPHE, 2001). The study area is marked by a rectangle where elevated As concentrations were observed to increase with distance from the river. (b) Google Earth image of the study area where elevated As levels (n = 913) measured with a field kit are clustered near the base of the Naga Patkai Hill ranges. Location of nine drilled boreholes (marked as triangles) along a 35-km transect (dashed line) is also shown. Symbol color corresponds to As concentration ≤10 (blue), 10–50 (green), and >50 μg/L (red), respectively.

from Assam, as well as the opposite pattern of As concentrations declining with distance from the river documented further downstream in Bangladesh, are actually controlled by the same set of processes.

2. Study Site and Methods

2.1. Study Site

The study area is an alluvial plain in the upper Brahmaputra Valley bounded by the Brahmaputra River to the north, the Mikir Hills to the west, and the Naga Patkai Hill ranges to the south (Figure 1). The main geomorphic units within this area are the floodplain of the Brahmaputra River in the north, a central upland area covering younger and older alluvial plains, and the piedmont plains to the south (CGWB, 2008). In addition to the Brahmaputra River, the fluvial sediments are supplied by two southern tributaries, the Dhansiri and Bhogdoi Rivers, which drain the Naga Patkai Hill ranges and Mikir Hills of Quaternary age (Evans, 1932; Mathur & Evans, 1964). Shifts in the course of these rivers have been attributed to neotectonism (Sharma, 2005). Reports from previous drilling in the area to a depth of 400 m indicate a thick, multilayered aquifer system composed of medium- to coarse-grained sand (CGWB, 2008). Available lithologs indicate that the aquifer is capped to the east by a 30–50-m-thick layer of clay and to the west by clay layers of more variable thickness.

2.2. Arsenic Measurements With Field Kit

The Arsenic Econo-Quick test kit (Part No. 481298, Industrial Test Systems Inc., Rock Hill, SC, USA) was used to test a total of 777 private and public wells <60-m deep within a ~2,000-km^2 area between June and August

2015 and an additional 136 wells closer to the drilling transect in November 2015. Groundwater samples were collected after hand pumping for 3–5 min to purge each well. The kit relies on the nineteenth century Gutzeit method, which is based on the generation of arsine gas from an acidified sample to which zinc powder is added. The arsine is trapped on and changes the color of a suspended test strip impregnated with mercuric bromide. The entire procedure requires less than 15 min. The color of the test strip is compared by eye with a reference chart corresponding to nine bins of As concentrations in the 0–1,000-μg/L range. The oxidizing reagent ("Reagent 2") was not used because no hydrogen sulfide levels in the well water sufficient to cause signal suppression were detected by smell. George et al. (2012) showed that kit measurements were consistent with laboratory measurements for well water from Bangladesh, provided that a systematic twofold overestimate of As concentrations above 50 μg/L by the kit is taken into account. Well depths recorded while testing are based on the household's recollection, which is typically correct because it has witnessed every section of PVC it paid for to go into the ground at the time of installation. The same kit was used to determine the effectiveness for As of a subset of 22 of the home-made iron removal filters that are used by most households in the area. These devices consist of a plastic or metal container partially filled with sand and a cloth filter and a hole at the bottom (Figure S1 in the supporting information).

2.3. Sediment and Groundwater Sampling

Sediment cuttings were collected from nine boreholes drilled along a 35-km transect perpendicular to the Brahmaputra River (Figure 1b). Local drillers used the indigenous manual "hand-flapper" or "sludger" method for installing wells (Horneman et al., 2004). Cuttings were collected at 5-ft (1.5-m) intervals to a depth of up to 60 m and, in the case of sandy intervals, washed off their clay content using clean local well water to minimize the contribution from recycled drilling water. A pressed pellet of moist sediment from each interval was wrapped in transparent cling wrap for archiving. At three of the drill sites located 5, 19, and 41 km from the river, respectively, monitoring wells screened at 45, 32, and 57-m depth were installed.

The field kit was also used to estimate in the field the exchangeable fraction of As in aquifer sands. About 0.5 g of freshly collected and washed sand was added to the kit's reaction vessel along with 50 mL of (low-As) bottled water. The slurry was processed as a water sample (again without Reagent 2), and a visual reading of the strip against the reference chart was obtained after the standard 10-min reaction time.

2.4. Sediment Reflectance

A CM700d diffuse reflectance spectrophotometer (Minolta Corp., USA) was used to quantify changes in the color of the freshly collected clay and sand cuttings through the cling wrap at the drill site. The instrument was calibrated each day with the white barium sulfate plate provided by the manufacturer. Readings corresponded to an illuminant source of D65 (daylight-containing ultraviolet component, color temperature 6504 K), with the observer angle set to 10°, and excluding the specular component (direct reflection). Previous work has shown that the proportion of Fe (II)/Fe in the acid-leachable fraction of Fe oxides in aquifer sediments of the Bengal Basin is linearly related to the difference in reflectance between 530 and 520 nm (Horneman et al., 2004). The drift in reflectance difference at these wavelengths over the course of the day for the calibration plate remained $\leq$0.01%.

2.5. Bulk Sediment Composition

Both clay and sand cuttings were also analyzed at the drill site for bulk composition through the cling wrap with an InnovX Delta Premium hand-held X-ray fluorescence analyzer mounted upside down in a portable stand. The samples were analyzed in the "Soil" mode, for a total duration of 180 s at the instrument's three incident X-ray energies. The calibration of the instrument was checked against National Institute of Standards and Technology (NIST) reference material SRM 2711 (Montana soil) at the beginning and the end of each day. A total of 14 readings for this standard over the course of the field work averaged 111 $\pm$ 6 mg/kg relative to a certified value of 105 $\pm$ 8 mg/kg. No correction was made to adjust for the coarse grain size of the sand cuttings.

2.6. Arsenic Measurements in the Laboratory

To verify the performance of the kit in a groundwater matrix that may be different in Assam compared to Bangladesh, groundwater was collected without filtration in 20-mL polyethylene scintillation vials (Wheaton no. 986706, with PolySeal-lined cap) from a subset of 288 wells. One week prior to analysis by high-resolution inductively coupled plasma mass spectrometry (ICP-MS) on a Thermo-Finnigan Element2 (Cheng et al., 2004),

the samples were acidified to 1% Optima HCl. By comparing duplicate samples acidified in the field and in the laboratory, van Geen et al. (2007) have shown that this protocol ensures that any precipitated Fe oxyhydroxide redissolves. The advantage of the method is that it eliminates the need for transporting concentrated HCl and reduces the chances of contamination. Samples of input and treated water from each of the 22 homemade iron removal systems were also analyzed by ICP-MS. An in-house consistency standard of artificial groundwater containing 430-μg/L As and reference materials NIST1640a (8.08 ± 0.07-μg/L As) and NIST1643e (58.98 ± 0.7-μg/L As) were included with every run to document accuracy and precision of the method to within <5%.

2.7. Radiocarbon Dating of Sediment and Groundwater

To constrain the age of the aquifer, a total of 13 clay samples from eight of the drill sites were selected for dating (no clay was encountered while drilling the site closest to the Brahmaputra River). The radiocarbon content of organic carbon in the clay samples was measured following standard procedures at the National Ocean Sciences Accelerator Mass Spectrometry Facility (NOSAMS) in Woods Hole, Massachusetts.

Groundwater samples were collected from the three monitoring wells for radiocarbon dating of dissolved inorganic carbon (DIC) in 250-mL glass bottles with Polyseal caps and poisoned with 0.2-mL of saturated $HgCl_2$. Ratios of $^{14}C/^{12}C$ and $^{13}C/^{12}C$ in DIC were also measured at NOSAMS following established procedures (Elder et al., 1998).

2.8. Groundwater Level Measurements

Groundwater levels in three monitoring wells along with nearby paired shallow wells were recorded manually once a month from March to September 2015 using a Solinst water-level meter. The three paired shallow wells were household tube wells selected as additional monitoring wells for a better representation of water level changes along the transect. The paired shallow wells (LAN2 at a distance of 500 m from monitoring well LAN1; BAT3 1,500 m from BAT1, and MAD2 500 m from MAD1) were the nearest available that the owner would allow us to open to lower the water level meter. In April 2018, groundwater levels were measured from an additional 13 wells screened in the 40–270-m depth range in an attempt to constrain the deeper portion of the groundwater flow model (Table S1). This second group of wells were either active or abandoned wells installed to feed Public Water Water Supply Schemes. For all active wells, pumping was stopped at least 3 hr before the water level was measured. Water level data were referenced to the extent possible to a common datum using Shuttle Radar Topography Mission (SRTM) elevation data for three 30 m × 30 m grid points closest to each of the monitoring wells. Ground elevation at each of the wells was estimated at 82 m (LAN, 5 km from the Brahmaputra River), 93 m (BAT, 19 km), and 98 m (MAD, 41 km) relative to sea level. The absolute uncertainty of SRTM elevations has been estimated elsewhere at ±16 m (Miliaresis & Paraschou, 2005), but the relative uncertainty appears to be considerably lower along the transect (Figure 2).

2.9. Groundwater Flow and Advective Transport Modeling

The U.S. Geological Survey flow code MODFLOW (Harbaugh, 2005) was used to compare various simplified 2-D cross-sectional models of the drilling transect. The modeled section is 50.5-km long with an average thickness of 400 m, approximating the thickness of the alluvium in this area. The model has a no-flow boundary at the bottom corresponding to the underlying basement rock (CGWB, 2013a, 2013b). Two different sets of models assume a no-flow boundary and a constant head boundary for the vertical edge of the model near the hills, respectively (Figure S2). Most of the model top is a recharge boundary with a constant topographic slope of 3 m/km. The only exception is the model top covering the half-width of the river, from river bank to the middle of the river, which is a constant head boundary equal to the annual average river stage of 74 m at this location. Another no-flow vertical boundary is placed at the river end of the model in the middle of the Brahmaputra River reflecting symmetry of the flow system on either side of the river. The entire model section is divided into 83 rows and 202 columns, resulting in a total of 16,766 cells. Various versions of the model assume recharge rate along the transect, different vertical boundary condition assigned at the hillside boundary of the model, or both. In one version of the model, the transect was subdivided into three areas with different recharge rates that reflect variations in the thickness of clay overlying the aquifer. Additional model simulations consider plausible upward and downward hydraulic head gradients along the foothill boundary of the model and different pumping scenarios based on the available information. All simulations were run for steady state conditions.

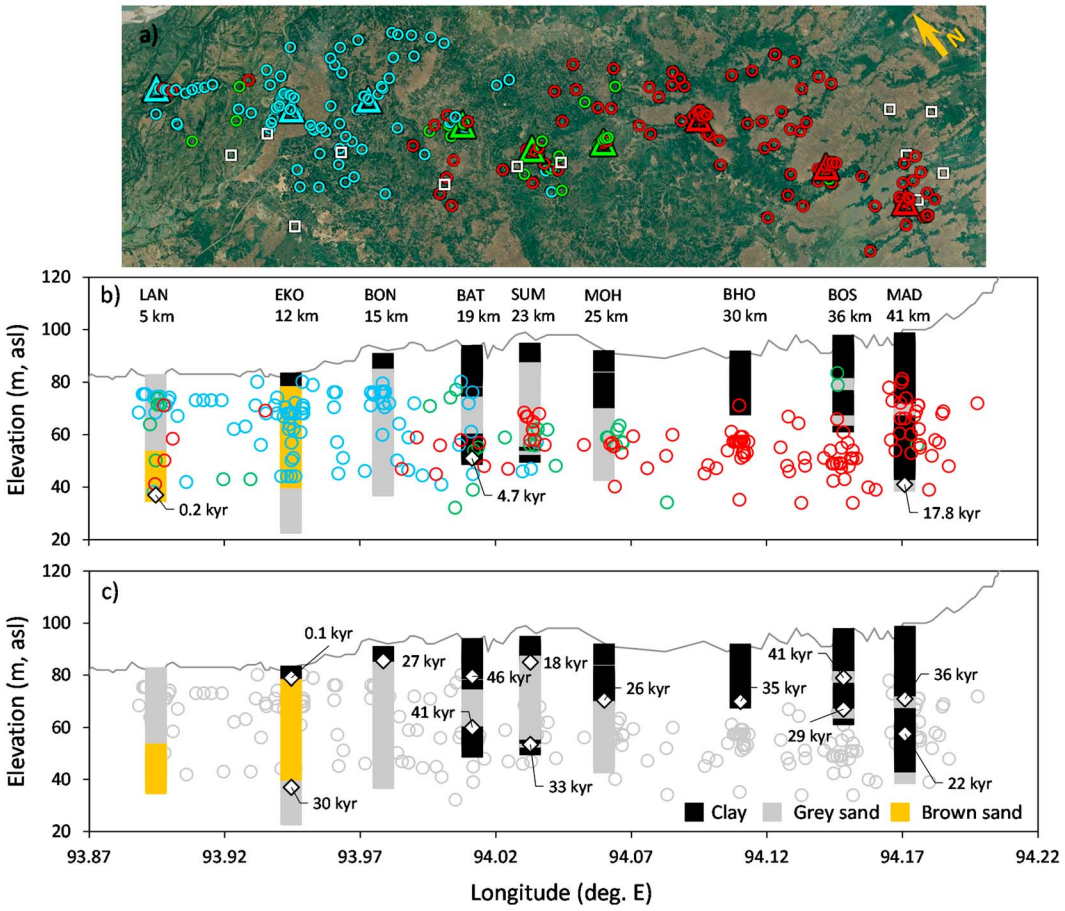

Figure 2. (a) Map showing As concentrations in shallow wells (<60-m depth) along the 35-km transect (n = 242). Location of drilled boreholes marked by triangles labeled as in Table 2. Symbol color in (a) and (b) shows As concentrations in groundwater (blue ≤10 μg/L, green 10–50 μg/L, and red >50 μg/L). White squares show the location of wells listed in Table S1. (b) Depth distribution of As in groundwater along the transect. Radiocarbon ages for DIC in groundwater from three monitoring wells marked as diamonds are also shown. (c) Lithology inferred from boreholes drilled along the transect shown with radiocarbon ages for organic carbon contained in clay samples marked as diamonds. Gray open circles in (c) correspond to the same wells as in (b). Gray full line in (b) and (c) represents land surface elevation of the transect line inferred from SRTM. Note: asl denotes above sea level.

Groundwater ages were simulated by forward particle tracking using the United States Geological Survey (USGS) particle transport code MODPATH (Pollock, 2012). For the model with a no-flow boundary at the hillside, nine particles of zero age were placed at each model cell along the model top boundary (a recharge boundary) and tracked forward. For the model with a constant head boundary, nine additional particles per cell with an initial age of 18 kyr were placed near the hills and tracked in forward direction. In this case, the assumption is that the water at this location is recharged at distant locations and therefore already old.

3. Results

3.1. Arsenic in Groundwater

Field kit data for a total of 913 wells <60-m deep partially covering the districts of Golaghat and Jorhat indicate that 33% of the wells in the area met the WHO guideline of 10 μg/L for As in drinking water, another 21% contained >10 to 50 μg/L, and 46% of samples exceeded 50 μg/L (Figure 1b). The vast majority of wells within 10 km of the main course of the Brahmaputra contain <10 μg/L for As, whereas most wells within 10 km of the Naga foothills contain >50 μg/L. Reported well depths near the river and the Naga foothills ranged 5 to 40 m and 20 to 60 m, respectively. Correcting for a 20-m difference in elevation from the Brahmaputra River to the Naga foothills, the screens of the vast majority of wells tested within a distance of 5 km of the transect therefore span the 40–80-m range in elevation (Figures 2a and 2b).

Table 1
Comparison of As Concentrations Measured in the Field Using Arsenic Quick™ Field Test Kit (ITS, Rock Hill, SC, USA) and the Laboratory Using ICP-MS

		Field data		
	As	0–10 μg/L	25–50 μg/L	100–1,000 μg/L
Lab data	≤10 μg/L	84 (99%)	5	0
	>10–50 μg/L	1	27 (47%)	13
	>50 μg/L	0	26	131 (91%)

Note. The proportion of wells correctly classified by the Arsenic Quick™ field test kit indicated in parentheses is relative to the sum in each column.

Comparison of field and laboratory data for a subset of 288 wells confirms that the kit results are by and large consistent with the ICP-MS measurements. Samples giving kit readings of 0–10 μg/L, which averaged 3 ± 6 μg/L by ICP-MS, and > 50 μg/L (120 ± 90 μg/L) misclassified 1% and 9% of wells, respectively (Table 1). A larger proportion of 53% of wells in the intermediate category with readings of 25–50 μg/L were incorrectly classified, but not a single well with >50 μg/L was classified by the kit as containing ≤10 μg/L and vice versa. Relative to the WHO guideline of 10 μg/L, the kit misclassified a total of only 6 (2%) out of all 288 tested wells (Figure S3). The ICP-MS data also confirm that the kit overestimates As concentrations about a factor of 2 at concentrations >50 μg/L in Assam, as previously reported for wells in the Bengal Basin (Figure S3).

The input water from 18 of the 22 selected wells whose water was treated with a home-made device ranged from 51 to 280-μg/L As (Figure S4). The removal efficiency ranged from 36 to 92%, and the treated water of 12 of these 18 wells contained <50-μg/L As. However, the treated water from only 4 of the same 18 wells with >50 μg/L met the WHO guideline of 10 μg/L for As in drinking water.

3.2. Lithology and Redox State of Aquifer Sands

The borehole lithology shows a marked increase in the thickness of a surface clay layer starting from none to <5 m at the three sites closest to the river to 60 m of almost continuous clay at the most distant site near the base of the Naga foothills (Figure 2b). Near the river, fine- to medium-grained sands dominate, while only thinner lenses of sands were encountered in boreholes closer to the hills. No sand was recovered during drilling to 25-m depth at the third closest site to the Naga Hills. Most of the sand across the transect are gray, with the exception of the site closest to the river where brown sand was recovered between 20 and 45-m depth and the next closest site with brown sand extending from 5 to 45-m depth (Figure 2b). The reflectance data indicate that particularly reduced Fe oxides with a difference in reflectance <0.1% (Horneman et al., 2004) prevail in the middle portion of the transect. In contrast, less reduced gray sands occur above the layer of brown sand at the site closest to the river and as thinner sand layers at two sites closer to the Naga Hills (Figure 3a). Most clay samples along the transect are also gray, with the exception a few orange-brown clays at the site closest to the river and near the Naga Hills (Figure S5).

3.3. Composition of Aquifer Sands

Under the counting time selected for field analysis by XRF, the concentrations of As in as many as 121 of the total of 147 samples of sand cuttings that were analyzed was below the limit of detection of 1.4 to 1.8 mg/kg calculated by the manufacturer's software. For display, these readings were converted to concentrations corresponding to half the limit of detection and error bars extending from zero to the limit of detection (Figure 3b). Concentrations of As in the remaining samples range from 1.5 to 4.6 mg/kg, without any clear pattern distinguishing drill sites close to the Brahmaputra River and the Naga Hills, respectively. Concentrations of As in 23 clay samples were below the limit of detection of 1.3 to 2.4 mg/kg, while the remaining 92 samples range from 1.6 to 27.7-mg/kg As, and again do not show a clear pattern across drill sites (not shown).

Analyzing sandy cuttings with the field kit resulted in readings limited to 0 and 10-μg/L As for all 49 samples of sand cuttings from the five sites closest to the river (Table 2). The corresponding range in leachable As concentrations is 0–1 mg/kg without adjustment in the reference chart. Out of the nine samples of sand cuttings

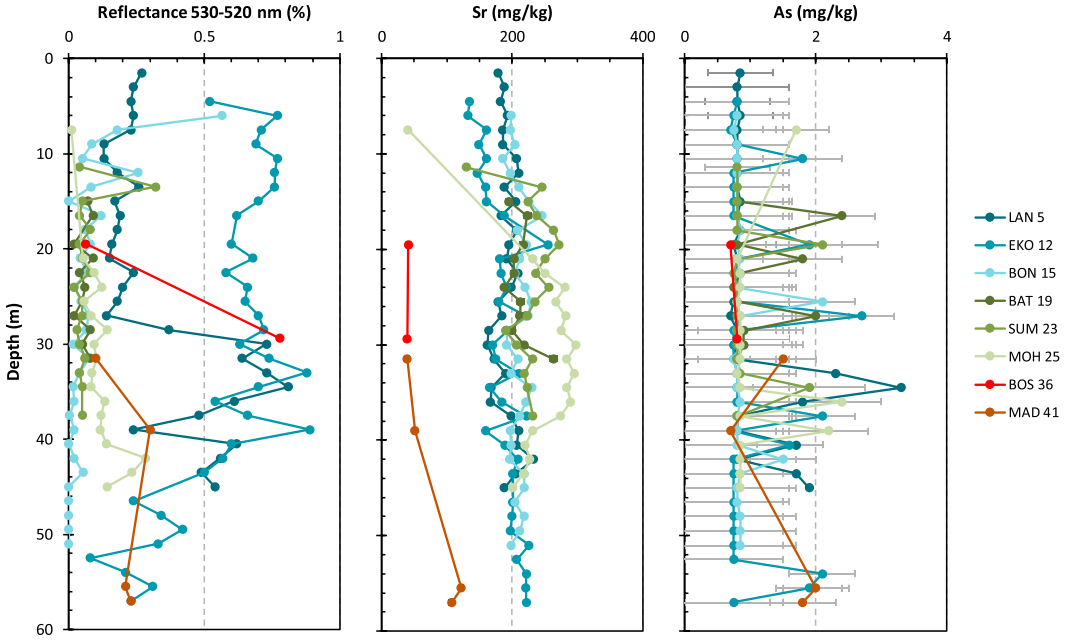

Figure 3. Profiles of properties of sand cuttings recovered from eight of the nine drill sites. (a) Difference in reflectance at 530 and 520 nm as proxy for sand color and the degree of reduction of Fe oxides (left = gray/reduced, right = brown/oxidized), concentrations of (b) Sr, and (c) As. Numbers next to each drilling site code indicate distance from the Brahmaputra River in kilometers.

analyzed from the four sites closest to the Naga Hills instead, three samples gave readings of 25 and 50 μg/L corresponding to 2.5–5-mg/kg leachable As.

With the exception of Sr, there was no marked difference in the bulk composition of aquifer sands for other elements (K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Rb, Zr, Ba, and Pb) that were measured by XRF. Concentrations of Sr in all but one sample of sand cuttings from the six sites closest to the Brahmaputra River ranged from 130 to 300 mg/kg (Figure 3c). In contrast, the Sr content of all 10 sand samples from the two sites closest Naga Hills was consistently lower and ranged from 50 to 120 mg/kg.

3.4. Radiocarbon Ages

With the exception of one very shallow sample at 0.3-m below ground level (bgl), the second closest site to the river, the uncorrected radiocarbon ages of organic matter in all clay cuttings range from 18 to 46 kyr (Figure 2c and Table S2). At three of the four sites dated at more than one interval, the radiocarbon age of the deeper interval is lower than for the shallower interval. At BAT, the age reversal could be because of ages 41 and 46 kyr are close to the practical limit for radiocarbon dating but this is not the case at BOS and MAD. Every effort was made to minimize the likelihood of shallow sediment falling into the hole while drilling, and only samples contained within large chunks of clays were selected for dating. The age reversals closest to the foothills may therefore indicate the occasional transport and deposition of older reworked material. Overall, the data unambiguously indicate pre-Holocene (>12-kyr) deposition along most of the transect. The uncorrected DIC ages for groundwater span a wide range from 0.2 kyr at the site closest to the river to 4.7 kyr near the middle of the transect and 17.8 kyr closest to the Naga Hills (Figure 2b and Table S2).

Table 2
Arsenic Concentrations Measured With the Kit in Slurries of Sand Cuttings at Nine Drill Sites

Borehole ID	Distance from river (km)	As (μg/L)			
		0	10	25	50
		No. of samples			
LAN	5	8	5	0	0
EKO	12	16	0	0	0
BON	15	6	0	0	0
BAT	19	3	3	0	0
SUM	23	2	6	0	0
MOH	25	0	2	1	0
BHO	30	0	0	0	0
BOS	36	1	0	0	0
MAD	41	1	2	0	2

3.5. Groundwater Levels and Head Gradient

Groundwater levels range across the transect from 2 to 10 m below the local ground surface (Table 3). When referred to a constant datum,

Table 3
Variations in Water Level Data Relative to the Local Ground Surface for Three Installed Monitoring Wells and Three Shallower Existing Wells Along the Transfect

Well ID	Coordinates	Depth (m)	Elev. (m)	Water level (m)					
				Feb	Mar	Apr	May	Jun	Sep
LAN1 inst. well	26.7345°N; 93.8947°E	45	82	3.25	3.5	3.55	3.42	1.37	1.7
LAN2 exist. well	26.7350°N; 93.8953°E	8	82	3.2	3.35	3.37	3.35	1.32	1.76
BAT1 inst. well	26.6510°N; 94.0114°E	32	93	7.8	7.6	7.62	7.6	7.47	7.58
BAT3 exist. well	26.6525°N; 94.0123°E	8	93	6.1	6.3	6.35	6.3	5.87	5.45
MAD1 inst. well	26.5212°N; 94.1709°E	57	98	8	9.7	9.75	9.7	7.2	7.87
MAD2 exist. well	26.5230°N; 94.1694°E	26	98	2.4	2.5	2.5	2.5	2.4	2.92

Note. Also listed are well depths relative to the local ground surface and estimated ground elevation based on SRTM.

shallow groundwater elevations decrease from 95 m near the hills to 80 m at the river, corresponding to a lateral head gradient of 0.3×10^{-3} (Figure 4). Whereas there is no detectable vertical groundwater head gradient between the shallow and deeper well monitored closest to the river (LAN at 5 km), the groundwater level was about 2 and 5 m lower in the deeper well at BAT (19 km) and MAD (41 km) than shallower wells, respectively. Groundwater levels shoaled by about 1–2 m from March–May to July–September in three of the monitored wells but remained essentially constant in the other wells.

Groundwater levels measured in mostly deeper wells along the transect in 2018 are not contemporaneous with the shallow head measurements but can still be used to place some limits on possible vertical head gradients. Taking into account the considerable uncertainty in surface elevation, groundwater levels in the 50–80 m and 120–270 elevation range were within a few meters of each other (Figure S6) near the Naga Hills in 2018. Similarly, groundwater levels within the 10–80-m depth range measured in the 2015 and 2018 do not seem systematically different across the transect up to the Brahmaputra River.

3.6. Groundwater Flow Modeling

The groundwater modeling starts from the simplest case and then adds the minimum number of additional features needed to match the available observations. The case of the no-flow boundary near the hills implies that water in the aquifer originates solely from recharge across the top boundary. Constraints imposed by DIC radiocarbon ages are limited to the 35–50-m range in screen elevation, but groundwater head data are available for screens installed down to −174 m in elevation. Recharge in the study has been estimated by CGWB (2013a, 2013b) from seasonal water level changes and rainfall to be on the order of 250–300 mm/yr. For comparison, local precipitation averages 2,000 mm/yr. Pumping tests in the same area indicate hydraulic conductivities ranging from 40 to 90 m/d (Table S4). The lower end of the conductivity range, however, produces hydraulic heads that are much higher than observed assuming isotropy, particularly toward the foothills where the aquifer cannot accommodate an imposed recharge of 270 mm/yr (Figure S7a). By increasing the isotropic hydraulic conductivity to 80 m/d, the model reproduces more closely the observed heads for the same recharge (Figure S7b). A similar fit can be obtained by maintaining the hydraulic conductivity at 40 m/d but reducing recharge to 90 mm/yr (not shown). However, the simulated groundwater ages for all these cases are <5 kyr throughout the section and do not vary laterally (Figures S7a–S7d).

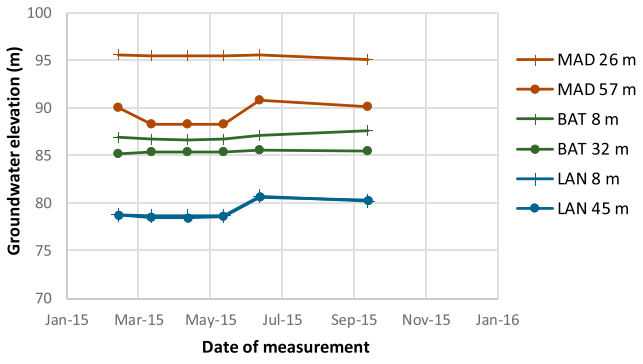

Figure 4. Temporal variability of groundwater elevation six monitoring wells along the transect.

Although simulated head gradients increase if the vertical hydraulic conductivity is reduced by a factor of 100–10,000 relative to the horizontal hydraulic conductivity, anisotropy does not markedly increase the groundwater ages (Figures S7c–S7e). Anisotropy on the order of 1,000 has previously been used to model the Ganges-Brahmaputra basin (Michael & Voss, 2008). In order to fit the observation more closely, and assuming Kh/Kv = 1,000, the recharge rate in Zones 2 and 1 was sharply reduced by a factor of 10 and 100 relative to Zone 3, respectively (Figure 5a). The model prediction is not particularly sensitive to the choice of anisotropy (Figures S7f and S7g). Groundwater ages still do not vary much laterally, and, significantly, the distribution

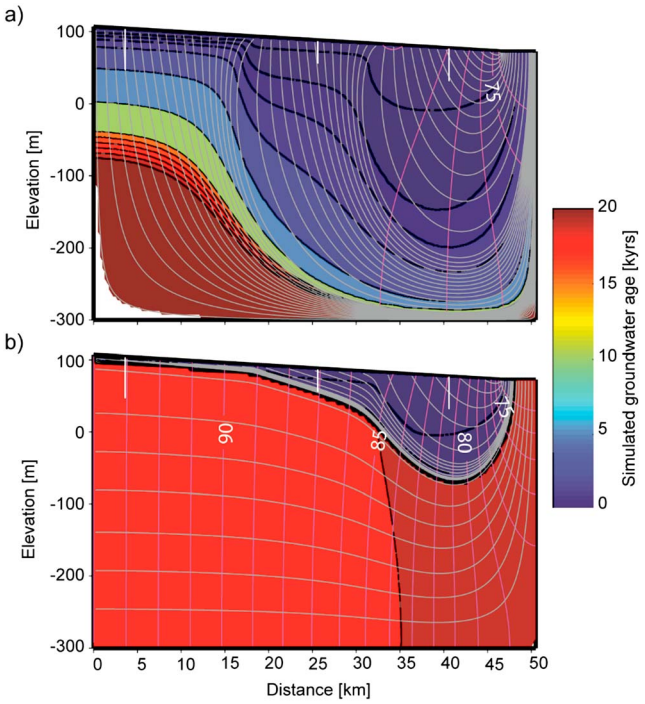

Figure 5. Simulated head and groundwater age distribution for (a) Case-1b-4: no-flow boundary at the hills (0 km); Kh = 80 m/d, and Kv = 0.08 m/d; recharge Zone 1 = 2.7 mm/yr, Zone 2 = 27 mm/yr, and Zone 3 = 270 mm/yr. (b) Case-2b3: constant head of 94 m at the hills (0 km) with 18-kyr initial age of water entering that boundary; Kh = 80 m/d and Kv = 0.08 m/d; recharge Zone 1 = 2.7 mm/yr, Zone 2 = 27 mm/yr, and Zone 3 = 270 mm/yr. Purple lines show head, and the gray lines show flow paths. Groundwater ages are shown by background color fill. The location and depths of the monitoring wells are shown as vertical white lines.

and range of simulated hydraulic gradients along the transect become inconsistent with the observations.

The model fit to both groundwater heads and ages improves considerably when using, instead, a constant head boundary near the hills (Figure 5b). The assumption is that water recharges far from the transect and flows a considerable distance before reaching it. The groundwater is therefore already 18-kyr old when it reaches the hillside boundary of the transect and ages further while flowing toward the river. At the same time, younger groundwater recharged from the top flows toward the river on top of the older groundwater. Because there is little or no clay cap near the river, recharge from the top pushes the old groundwater downward by creating a shallow flow system of young age water (<0.5 kyr); this is the base case scenario for subsequent model simulations. The basic pattern model prediction is again not very sensitive to the choice of anisotropy (Figures S7h and S7i); however, it is highly sensitive to the choice of spatial distribution of recharge (Figure S7o). Also, the distribution of groundwater ages and heads remains consistent with the observation if the lowest value of K's (Kh = 40 m/d, Kv = 0.04 m/d) obtained from pump test data is used (Figure S7p). The results are also not markedly different from the base case when either a downward decreasing or increasing head gradient of 4 m is introduced at the model boundary close to the Naga Hills (Figures S7j and S7k).

The base case responds to various plausible pumping scenarios but again not in a way that drastically alters the predicted distribution of heads and ages within the depth ranges where this information was collected. According to CGWB (2013a, 2013b), groundwater withdrawals in the region amount to approximately 60 mm/yr. When such pumping is drawn from the shallow aquifer, older water is drawn up but not in a way that drastically alters groundwater ages at the bottom of the monitoring wells (Figure S7l). In contrast, younger water is drawn down when pumping is imposed at greater depth again without drastically altering groundwater ages in the monitoring wells (Figure S7m). The impact on the distribution of groundwater ages is essentially undectable when pumping is imposed from depths above and below the screen of the monitoring wells (Figure S7n).

4. Discussion

4.1. Is Groundwater As Regulated by Bulk Sediment Geochemistry?

The new groundwater data are consistent with results for Golaghat and Jorhat districts of Assam previously reported by Mahanta et al. (2015) and Verma et al. (2016) but fill gaps in the coverage to produce a clear regional pattern of groundwater As concentrations steadily increasing from the banks of the Brahmaputra River to the Naga foothills (Figure 1b). The increase in groundwater As concentrations with distance from the river observed in Assam can only partially be explained by difference in the extent of reduction of Fe oxides in aquifer sands. Groundwater As levels are very low near sites LAN and EKO closest to the river, which contain intervals of oxidized brown sands (Figure 2b). Even the reflectance profile at LAN also indicates a sizeable layer of gray sands. Groundwater As concentrations are still generally low northwest of site BON, the third closest to the river, even though sands are uniformly gray at this location. The most telling indication that the redox state of aquifer sediments alone cannot account for variations in groundwater As are reflectance data showing particularly reduced sands prevailing in the middle portion of the transect instead of the region closest to the Naga Hills (Figure 3a).

Pickering et al. (2013) and Goodbred et al. (2014) have shown that bulk Sr concentrations can be used to distinguish Brahmaputra River sediments in Bangladesh derived from Sr-rich (140-mg/kg) Tibet and Sr-depleted

terranes of the Himalaya and Shillong Massif (<90 mk/kg). On this basis, a source other than Tibet can be assigned only to sands recovered from sites BOS and MAD closest to the Naga Hills (Figure 3b).

Bulk As concentrations in sand cuttings recovered during drilling are not related in any systematic way to groundwater As concentrations along the transect (Figure 3c). Whereas sites BOS and MAD cover the region where the groundwater As are most consistently elevated, many wells near the middle of the transect are also elevated in As. It therefore does not appear that the redox state, provenance, or the bulk As content of aquifer sands can explain the main trend of groundwater As concentrations across the transect.

4.2. Is Groundwater As Regulated by Hydrogeology?

In contrast to bulk As concentrations, concentrations of leachable As in the sediment estimated with the field kit do broadly follow that of groundwater As concentrations and increase from the Brahmaputra River toward the Naga Hills. Using a somewhat more involved method to displace As from adsorption sites, van Geen et al. (2008) showed that this type of relation between exchangeable As in Holocene sediment and groundwater As can be attributed to equilibrium exchange. Sizeable concentrations of exchangeable As in shallow gray sands have been confirmed by a series of push-pull experiments in Bangladesh during which low-As water was pumped into a high-As aquifer and vice versa (Radloff et al., 2017). The broad increase in both groundwater As concentrations and exchangeable As levels in gray sediment from the river to the hills can therefore plausibly be attributed to some form of equilibration, even if the sediment is of Pleistocene rather than Holocene age.

The pool of exchangeable As in aquifer sands is much larger than in groundwater. Under the assumption of equilibrium exchange, the pattern of groundwater As concentrations in the study area therefore requires a mechanism that explains the increase in exchangeable As in the solid phase toward the Naga Hills. The groundwater flow model that most closely matches the available data indicates that flushing over time could be the primary cause of the observed increase in exchangeable As and groundwater As from the Brahmaputra River to the Naga Hills. Orange sands are excluded from the argument because their Fe oxide coatings are insufficiently reduced to maintain a pool of exchangeable As (van Geen et al., 2004; Zheng et al., 2005). Near the river, the model indicates groundwater ages of only a few hundred years down to several tens of meters because of the permeability of the surface soils and proximity to the river discharge area (Figure 5b). The available As data suggest that the resulting flushing was sufficient to lower the amount of exchangeable As in these shallow Pleistocene sands. With increasing distance from the river, however, the age of groundwater, and therefore its residence time, increases much more rapidly with depth to several thousand years and higher (Figure 5b). This much more sluggish circulation, consistent with the radiocarbon ages, apparently preserved a larger pool of exchangeable As and, therefore, maintains a higher As concentrations in groundwater toward the Naga Hills. Various versions of the model show that the separation between a shallow rapid and a deep slower groundwater circulation system is not particularly sensitive to the choice of parameters or perturbation by pumping (Figure S7). Alternative explanations cannot be ruled out, but this is the simplest that is consistent with all the data and process studies conducted elsewhere in South Asia.

4.3. Arsenic Mitigation

Analysis of effluent from a limited number of home-made filters used to remove Fe from groundwater shows that As concentrations were brought down to <50 μg/L from higher levels in two third of cases, but <10 μg/L for only one quarter of these. Unlike in Vietnam, where Fe removal at the household level is particularly effective at removing As from groundwater because of relatively high Fe and low phosphate levels (Berg et al., 2006), additional measures providing access to low-As drinking water are clearly needed to lower the exposure of the rural population of Assam. Our survey included four wells >90-m deep (not shown) located near the study transect that met the WHO guideline value of 10 μg/L for As in drinking water. However, none of these are located within the area where most shallower wells are high in As.

In Bangladesh, the government's Department of Public Health Engineering has installed several hundred thousand deep wells throughout the country intended as public sources of low-arsenic drinking water (Ravenscroft et al., 2014), but the Assam Public Health Engineering Department has not conducted the drilling necessary to explore this approach. Instead, the APHED has installed over the past 40 years several thousand piped-water supply schemes to meet drinking water needs of the population, including 994 schemes in Golaghat and Jorhat districts that are connected to 3,548 delivery points (Integrated Management Information

System, 2017). The source water for these systems is typically groundwater processed through a Fe-based As removal plant where needed. The water is also chlorinated before delivery and tested for As by APHED district laboratories. Our testing of six such systems indicated As concentrations >50 μg/L in one case, two with 10–50 μg/L, and three containing <10-μg/L As. An alternative approach to treatment, at least in areas with mixed distribution of low- and high-As wells, might be to deliver through the existing pipes groundwater from existing wells that are already low in As. The time scale of the flushing that reduced exchangeable As levels in shallow aquifers from presumed high initial levels suggests that considerable volumes could be pumped from the low-As wells before drawing in As from adjacent contaminated aquifer.

The total population of Golaghat and Jorhat districts is 2 million, with an estimated 590,000 people living within the four blocks (Golaghat East and Gomariguri in Golaghat districts and Jorhat and Titabor in Jorhat districts) where the vast majority of tested wells (358 out of 481 wells tested) exceed 50-μg/L As, the permissible limit in the absence of alternative drinking water source in India (Bureau of Indian Standards, 2012). According to the Indian government's Ministry of the Drinking Water and Sanitation (Integrated Management Information System, 2017), these piped-water supply schemes provide full access to low-As water to 278,000 inhabitants within the four heavily As-contaminated blocks, and partial access to another 184,000 inhabitants, none to the remaining 128,000 inhabitants. Those villagers most in need will hopefully be targeted by additional water treatment systems in the planning with support from the Government of India and the World Bank.

5. Conclusions

The increase in shallow groundwater As concentrations with distance from the Brahmaputra River documented in this study is the opposite of the pattern observed downstream in Bangladesh. The underlying mechanism linking recharge and As in shallow aquifers, however, appears to be similar in the two regions. The combined set of observations indicates that surface permeability regulates the rate of recharge and flushing over centuries to thousands of years and, therefore, the level of As in shallow groundwater. Low-As aquifers could therefore probably be used more extensively as a source to reduce the exposure of villagers in the more affected portions of the region, at least in the short to medium term.

Acknowledgments

This study was supported by a grant from CE/C/CM/57 to C. M., a Fulbright fellowship to R. C., U.S. National Institute of Environmental Health Sciences grant P42 ES10349, and US National Science Foundation grant NSF ICER 1414131. Key data collected under this project are provided as tables in the text and supporting information. Additional data are archived at http://www.hydroshare.org/resource/bbe23dfacab647568a18dc338114d6d7. We are immensely grateful to Ubadur Rahman Neog, Abidur Rahim, and Dilip Kakoti from the Assam Public Health Engineering Department for their generous support and collaborative spirit provided throughout the project. We also thank students from IIT Guwahati, Dr. Chander Kumar Singh from TERI Institute for Advanced Studies, and Mr. Imtiaz Choudhury from the University of Dhaka for their help in the field. This is Lamont-Doherty Earth Observatory contribution no. 8238.

References

Ahmed, K. M., Bhattacharya, P., Hasan, M. A., Akhter, S. H., Alam, S. M. M., & Bhuyian, M. A. H. (2004). Arsenic contamination in groundwater of alluvial aquifers in Bangladesh: An overview. *Applied Geochemistry*, *19*(2), 181–200. https://doi.org/10.1016/j.apgeochem.2003.09.006

Assam Public Health Engineering Department (2017). Water quality problem. Retrieved from http://aphe.gov.in/waterqualityprblm.aspx. Accessed on 29 Oct, 2017.

Aziz, Z., van Geen, A., Versteeg, R., Horneman, A., Zheng, Y., Goodbred, S. J. Jr., & Ahmed, K. M. (2008). Impact of local recharge on arsenic concentrations in shallow aquifers inferred from the electromagnetic conductivity of soils in Araihazar, Bangladesh. *Water Resources Research*, *44*, W07416. https://doi.org/10.1029/2007WR006000

Berg, M., Luzi, S., Trang, P. T. K., Viet, P. H., Giger, W., & Stuben, D. (2006). Arsenic removal from groundwater by household sand filters: Comparative field study, model calculations, and health benefits. *Environmental Science & Technology*, *40*(17), 5567–5573. https://doi.org/10.1021/es060144z

Berg, M., Tran, H. C., Nguyen, T. C., Pham, H. V., Schertenleib, R., & Giger, W. (2001). Arsenic contamination of groundwater and drinking water in Vietnam: A human health threat. *Environmental Science & Technology*, *35*(13), 2621–2626. https://doi.org/10.1021/es010027y

BGS/DPHE (2001). Arsenic contamination of groundwater in Bangladesh. In D. G. Kinniburgh & P. L. Smedley (Eds.), *BGS Technical Report WC/00/19 British Geological Survey* (p. 267). Keyworth, UK.

Bhattacharya, P., Chatterjee, D., & Jacks, G. (1997). Occurrence of arsenic-contaminated groundwater in alluvial aquifers from Delta Plains, eastern India: Options for safe drinking water supply. *International Journal of Water Resources Development*, *13*(1), 79–92. https://doi.org/10.1080/07900629749944

Borah, K., Bhuyan, B., & Sarma, H. P. (2009). Lead, arsenic, fluoride, and iron contamination of drinking water in the tea garden belt of Darrang district, Assam, India. *Environmental Monitoring and Assessment*, *169*(1-4), 347–352. https://doi.org/10.1007/s10661-009-1176-2

Bureau of Indian Standards (2012). Drinking water—Specification, IS 10500:2012, Second revision ICS 13.060.20. Retrieved from http://cgwb.gov.in/Documents/WQ-standards.pdf. Accessed on 20 Oct, 2017.

CGWB (2008). Central Ground Water Board, Ministry of Water Resources Government of India, Information Booklet. Jorhat District. Retrieved from http://cgwb.gov.in/

CGWB (2013a). Ground Water Information Booklet Jorhat District, Assam, Ministry of Water Resources, Government of India, Guwahati, India.

CGWB (2013b). Ground Water Information Booklet Golaghat District, Assam, Ministry of Water Resources, Government of India, Guwahati, India.

Chakraborti, D., Sengupta, M. K., Rahman, M. M., Ahmed, S., Chowdhury, U. K., & Hossain, M. A. (2004). Groundwater arsenic contamination and its health effects in the Ganga–Meghna–Brahmaputra plain. *Journal of Environmental Monitoring*, *6*(6), 74N–83N.

Cheng, Z., Zheng, Y., Mortlock, R., & van Geen, A. (2004). Rapid multi element analysis of groundwater by high-resolution inductively coupled plasma mass spectrometry. *Analytical and Bioanalytical Chemistry*, *379*(3), 512–518. https://doi.org/10.1007/s00216-004-2618-x

Chetia, M., Chatterjee, S., Banerjee, S., Nath, M. J., Singh, L., Srivastava, R. B., & Sarma, H. P. (2011). Groundwater arsenic contamination in the Brahmaputra river basin: A water quality assessment in the Golaghat (Assam), India. *Environmental Monitoring and Assessment*, *173*(1-4), 371–385. https://doi.org/10.1007/s10661-010-1393-8

Choudhury, R., Sharma, P., Mahanta, C., & Sarma, H. P. (2015). Evaluation of the processes controlling arsenic contamination in parts of the Brahmaputra floodplains in Assam, India. *Journal of Environmental Earth Science*, *73*(8), 4473–4482. https://doi.org/10.1007/s12665-014-3735-2

Datta, S., Neal, A. W., Mohajerin, T., Ocheltree, T., Rosenheim, B. E., White, C. D., & Johannesson, K. H. (2011). Perennial ponds are not an important source of water or dissolved organic matter to groundwaters with high arsenic concentrations in West Bengal, India. *Geophysical Research Letters*, *38*, L20404. https://doi.org/10.1029/2011GL049301

Donselaar, M. E., Bhatt, A. G., & Ghosh, A. K. (2017). On the relation between fluvio-deltaic flood basin geomorphology and the wide-spread occurrence of arsenic pollution in shallow aquifers. *Science of the Total Environment*, *574*, 901–913. https://doi.org/10.1016/j.scitotenv.2016.09.074

Elder, K. L., Mcnichol, A. P., & Gagnon, A. R. (1998). Reproducibility of seawater, inorganic and organic carbon ^{14}C results at NOSAMS. *Radiocarbon*, *40*(01), 223–230. https://doi.org/10.1017/S0033822200018087

Evans, P. (1932). Tertiary succession in Assam. *Transactions of Mineralogical and Geological Institute, India*, *5*, 155–260.

Fendorf, S., Michael, H. A., & van Geen, A. (2010). Spatial and temporal variations of groundwater arsenic in South and South East Asia. *Science*, *328*(5982), 1123–1127. https://doi.org/10.1126/science.1172974

George, C. M., Zheng, Y., Graziano, J. H., Rasul, S. B., Hossain, Z., Mey, J. L., & van Green, A. (2012). Evaluation of an arsenic test kit for rapid well screening in Bangladesh. *Environmental Science and Technology*, *46*(20), 11,213–11,219. https://doi.org/10.1021/es300253p

Goodbred, S. L., Paolo, P. M., Ullah, M. S., Pate, R. D., Khan, S. R., Kuehl, S. A., et al. (2014). Piecing together the Ganges-Brahmaputra-Meghna River delta: Use of sediment provenance to reconstruct the history and interaction of multiple fluvial systems during Holocene delta evolution. *Geological Survey of America Bulletin*, *126*(11-12), 1495–1510. https://doi.org/10.1130/B30965.1

Goswami, R., Rahman, M. M., Murrill, M., Sarma, K. P., Thakur, R., & Chakraborti, D. (2013). Arsenic in the groundwater of Majuli – The largest river island of the Brahmaputra: Magnitude of occurrence and human exposure. *Journal of Hydrology*, *518*, 354–362. https://doi.org/10.1016/j.jhydrol.2013.09.022

Harbaugh, A. W. (2005). MODFLOW-2005, the US Geological Survey modular ground-water model-the ground-water flow process. U.S. Geological Survey Techniques and Methods 6–A16.

Hoque, M. A., Khan, A. A., Shamsudduha, M., Hossain, M. S., Islam, T., & Chowdhury, S. H. (2009). Near surface lithology and spatial variation of arsenic in the shallow groundwater: Southeastern Bangladesh. *Environmental Geology*, *56*(8), 1687–1695. https://doi.org/10.1007/s00254-008-1267-3

Horneman, A., van Geen, A., Kent, D. V., Mathe, P. E., Zheng, Y., Dhar, Y., et al. (2004). Decoupling of As and Fe release to Bangladesh groundwater under reducing conditions. Part I: Evidence from sediment profiles. *Geochimica et Cosmochimica Acta*, *68*(17), 3459–3473. https://doi.org/10.1016/j.gca.2004.01.026

Integrated Management Information System (2017). Number of habitations with piped water supply schemes district wise. Ministry of Drinking Water & Sanitation, National Rural Drinking Water Programme. Retrieved from http://indiawater.gov.in/IMISReports/Reports/BasicInformation/rpt_RWS_HabitationCoveredWithPWS_D.aspx?Rep=0. Accessed on 20 Oct 2017.

Mahanta, C., Choudhury, R., Basu, S., Hemani, R., Dutta, A., Barua, P. P., & Saikia, L. (2015). Preliminary assessment of arsenic distribution in Brahmaputra River basin of India based on examination of 56,180 public groundwater wells. In *Safe and sustainable use of arsenic-contaminated aquifers in the Gangetic Plain* (pp. 57–64). Guwahati, Assam: Springer Publishers.

Mailloux, B. J., Trembath-Reichert, E., Cheung, J., Watson, M., Stute, M., Freyer, G. A., et al. (2013). Advection of surface-derived organic carbon fuels microbial reduction in Bangladesh groundwater. *Proceedings of the National Academy of Sciences U.S.A.*, *110*, 5331–5335. https://doi.org/10.1073/pnas.1213141110

Mathur, L. P., & Evans, P. (1964). Oil in India. 2nd Int. Geol. Congr., New Delhi. p.85.

McArthur, J. M., Banerjee, D. M., Hudson-Edwards, K. A., Mishra, R., Purohit, R., Ravenscroft, P., et al. (2004). Natural organic matter in sedimentary basins and its relation arsenic in anoxic ground water: The example of West Bengal and its worldwide implications. *Applied Geochemistry*, *19*(8), 1255–1293. https://doi.org/10.1016/j.apgeochem.2004.02.00

McArthur, J. M., Nath, B., Banerjee, D. M., Purohit, R., & Grassineau, N. (2011). Palaeosol control on groundwater flow and pollutant distribution: The example of arsenic. *Environmental Science and Technology*, *45*(4), 1376–1383. https://doi.org/10.1021/es1032376

McArthur, J. M., Ravenscroft, P., Banerjee, D. M., Milsom, J., Hudson-Edwards, K. A., Sengupta, S., et al. (2008). How paleosols influence groundwater flow and arsenic pollution: A model from the Bengal Basin and its worldwide implication. *Water Resources Research*, *44*, W11411. https://doi.org/10.1029/2007WR006552

Meharg, A. A., Lombi, E., Williams, P. N., Scheckel, K. J., Feldmann, J., Raab, A., et al. (2008). Speciation and localization of arsenic in white and brown rice grains. *Environmental Science and Technology*, *42*(4), 1051–1057. https://doi.org/10.1021/es702212p

Michael, H. A., & Voss, C. I. (2008). Evaluation of the sustainability of deep groundwater as an arsenic-safe resource in the Bengal Basin. *Proceedings of the National Academy of Sciences of the United States of America*, *105*(25), 8531–8536. https://doi.org/10.1073/pnas.0710477105

Miliaresis, G. C., & Paraschou, C. V. E. (2005). Vertical accuracy of the SRTM DTED level 1 of Crete. *International Journal of Applied Earth Observation and Geoinformation*, *7*(1), 49–59. https://doi.org/10.1016/j.jag.2004.12.001

Nath, B., Berner, Z., Basu Mallik, S., Chatterjee, D., Charlet, L., & Stueben, D. (2005). Characterization of aquifers conducting groundwaters with low and high arsenic concentrations: A comparative case study from West Bengal, India. *Mineralogical Magazine*, *69*(05), 841–854. https://doi.org/10.1180/0026461056950292

Nath, B., Mallik, S. B., Stuben, D., Chatterjee, S., & Charlet, L. (2010). Electrical resistivity investigation of arsenic affected alluvial aquifers in West Bengal, India: Usefulness in identifying the areas of low and high groundwater arsenic. *Environment and Earth Science*, *60*(4), 873–884. https://doi.org/10.1007/s12665-009-0224-0

Neumann, R. B., Ashfaque, K. N., Badruzzaman, A. B. M., Ali, M. A., Shoemaker, J. K., & Harvey, C. F. (2010). Anthropogenic influences on groundwater arsenic concentrations in Bangladesh. *Nature Geoscience*, *3*(1), 46–52. https://doi.org/10.1038/ngeo685

Nickson, R. T., McArthur, J. M., Burgees, W., Ahmed, K. M., Ravenscroft, P., & Rahmanñ, M. (1998). Arsenic poisoning of Bangladesh groundwater. *Nature*, *395*(6700), 338. https://doi.org/10.1038/26387

Pickering, J. L., Goodbred, S. L., Reitz, M. D., Hartzog, T. R., Mondal, D. R., & Hossain, M. H. (2013). Late Quaternary sedimentary record and Holocene channel avulsions of the Jamuna and old Brahmaputra River valleys in the upper Bengal delta plain. *Geomorphology*, *227*, 123–136. https://doi.org/10.1016/j.geomorph.2013.09.021

Polizzotto, M. L., Kocar, B. D., Benner, S. G., Sampson, M., & Fendorf, S. (2008). Near-surface wetland sediments as a source of arsenic release to groundwater in Asia. *Nature*, *454*(7203), 505–508. https://doi.org/10.1038/nature07093

Pollock, D. W. (2012). User guide for MODPATH version 6: A particle tracking model for MODFLOW—Techniques and methods 6–A41, U.S. Geological Survey.

Postma, D., Larsen, F., Thai, N. T., Trang, P. T. K., Jacobson, R., Nham, P. Q., et al. (2012). Groundwater arsenic concentrations in Vietnam controlled by sediment age. *Nature Geoscience*, *5*(9), 656–661. https://doi.org/10.1038/ngeo1540

Radloff, K. A., Zheng, Y., Stute, M., Weinman, B., Bostick, B., Mihajlov, I., et al. (2017). Reversible adsorption and flushing of arsenic in a shallow, Holocene aquifer of Bangladesh. *Applied Geochemistry*, *77*, 142–157. https://doi.org/10.1016/j.apgeochem.2015.11.003

Ravenscroft, P., Brammer, H., & Richards, K. (2009). *Arsenic pollution: A global synthesis*, *RGS-IBG Book Series*, Chichester, UK: Wiley-Blackwell. https://doi.org/10.1002/9781444308785

Ravenscroft, P., Kabir, A., Hakim, S. A. I., Ibrahim, A. K. M., Ghosh, S. K., Rahman, M. S., et al. (2014). Effectiveness of public rural water points in Bangladesh with special reference to arsenic mitigation. *Journal of Water Sanitation and Hygiene for Development*, *4*(4), 545–562. https://doi.org/10.2166/washdev.2014.038

Sharma, J. N. (2005). Fluvial process and morphology of the Brahmaputra River in Assam, India. *Geomorphology*, *70*(3-4), 226–256. https://doi.org/10.1016/j.geomorph.2005.02.007

Singh, A. K. (2004). Arsenic contamination in groundwater of north eastern India. In *Proceedings of 11th National Symposium on Hydrology with focal theme on water quality* (pp. 255–262). Roorkee: National Institute of Hydrology.

Smedley, P. L., & Kinniburgh, D. G. (2002). A review of the source, behavior and distribution of arsenic in natural waters. *Applied Geochemistry*, *17*(5), 517–568. https://doi.org/10.1016/S0883-2927(02)00018-5

Stuckey, J. W., Schaefer, M. V., Kocar, B. D., Benner, S. G., & Fendorf, S. (2016). Arsenic release metabolically limited to permanently water-saturated soil in Mekong Delta. *Nature Geoscince*, *9*(1), 70–76. https://doi.org/10.1038/ngeo2589

van Geen, A., Rose, J., Thoral, S., Garnier, J. M., Zheng, Y., & Bottero, J. Y. (2004). Decoupling of As and Fe release to Bangladesh groundwater under reducing conditions. Part II: Evidence from sediment incubations. *Geochimica et Cosmochimica Acta*, *68*(17), 3475–3486. https://doi.org/10.1016/j.gca.2004.02.014

Van Geen, A., Cheng, Z., Jia, Q., Seddique, A. A., Rahman, M. W., Rahman, M. M., & Ahmed, K. M. (2007). Monitoring 51 deep community wells in Araihazar, Bangladesh, for up to 5 years: Implications for arsenic mitigation. *Journal of Environmental Science and Health, Part A Toxic/Hazardous Substances and Environmental Engineering*, *42*(12), 1729–1740. https://doi.org/10.1080/10934520701564236

Van Geen, A., Zheng, Y., Goodbred, S. Jr., Horneman, A., Aziz, Z., Cheng, Z., et al. (2008). Flushing history as a hydrogeological control on the regional distribution of arsenic in shallow groundwater of the Bengal Basin. *Environmental Science and Technology*, *42*(7), 2283–2288. https://doi.org/10.1021/es702316k

Verma, S., Mukherjee, A., Mahanta, C., Choudhury, R., & Mitra, K. (2016). Influence of geology on groundwater-sediment interactions in varied arsenic enriched tectono-morphic aquifers of the Brahmaputra River basin. *Journal of Hydrology*, *540*, 176–195. https://doi.org/10.1016/j.jhydrol.2016.05.041

Weinman, B., Goodbred, S. L., Zheng, Y., Aziz, Z., Steckler, M., van Geen, A., et al. (2008). Contributions of floodplain stratigraphy and evolution to the spatial patterns of groundwater arsenic in Araihazar, Bangladesh. *Geological Society of America Bulletin*, *120*(11-12), 1567–1580. https://doi.org/10.1130/B26209.1

Winkel, L. H. E., Trang, P. T. K., Lan, V. M., Stengel, C., Amini, M., Ha, N. T., et al. (2011). Arsenic pollution of groundwater in Vietnam exacerbated by deep aquifer exploitation for more than a century. *Proceedings of the National Academy of Sciences U.S.A.*, *108*, 1246–1251. https://doi.org/10.1073/pnas.1011915108

Zheng, Y., van Geen, A., Stute, M., Dhar, R., Mo, Z., Cheng, Z., et al. (2005). Geochemical and hydrogeological contrasts between shallow and deeper aquifers in two villages of Araihazar, Bangladesh: Implications for deeper aquifers as drinking water sources. *Geochimica et Cosmochimica Acta*, *69*(22), 5203–5218. https://doi.org/10.1016/j.gca.2005.06.001