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Key Points:
- Groundwater >150 m deep is contaminated with arsenic in a 100-km-long latitudinal transect across the southwestern Bengal Basin
- Radiocarbon ages of deep groundwater are considerably lower in high-As area compared to the rest of the basin
- Modeling and analysis indicate that naturally deep flow in sand-dominated stratigraphy likely causes younger ages and high As concentrations

Supporting Information:
- Supporting Information S1

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High-Arsenic Groundwater in the Southwestern Bengal Basin Caused by a Lithologically Controlled Deep Flow System

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Abstract Elevated arsenic in Bengal Basin aquifers threatens human health. Most deep (>150 m) groundwater in Pleistocene aquifers is low in arsenic; however higher concentrations have been reported in the southwest border region. Here, we establish that this extensive arsenic contamination at depth is not associated with well failure. A combination of geochemistry and flow modeling constrains the factors that contribute to arsenic contamination at depth in this region. Deep groundwater in the affected area is younger (2.0 ± 0.6 kyr) than deep, low-arsenic groundwater elsewhere (12.0 ± 4.0 kyr) based on radiocarbon. Stratigraphic data indicate pre-Holocene deposition of the contaminated aquifers, but few low-permeability strata. Numerical modeling indicates that this stratigraphic anomaly permits a natural flow system that transports shallow groundwater to depth. Thus, in areas lacking low-permeability layers, arsenic contamination can occur in pre-Holocene aquifers and is probably not an early sign of future deep contamination in regions with interbedded low-permeability strata.

Plain Language Summary Exposure to arsenic in untreated groundwater pumped from millions of shallow wells across rural South Asia causes life-threatening cardiovascular disease and cancers in adults and reduces intellectual function in children. Deep (>150 m) groundwater low in arsenic is currently the most effective mitigation option in Bangladesh. This study shows that high concentrations of arsenic observed in deep aquifers in the border area between Bangladesh and India are likely confined to that region due to the absence of clay layers vertically dividing the sandy aquifer. Flow modeling constrained by groundwater dating shows that this contamination is therefore of natural origin and unlikely to expand to wider areas in the near future.

1. Introduction

Naturally occurring arsenic (As) concentrations in groundwater exceeding the World Health Organization drinking water guideline of 10 μg/l are found in aquifers on every continent (Coetsiers & Walraevens, 2006; Mandal & Suzuki, 2002; Ravenscroft et al., 2009; Smedley & Kinniburgh, 2002). In South and Southeast Asia, the problem is particularly severe because As concentrations in shallow, fluvio-deltaic aquifers are often orders of magnitude higher than the WHO guideline (Fendorf et al., 2010; Ravenscroft et al., 2005), and more than 100 million people in the Bengal, Mekong, and Red River deltas are at risk of adverse health effects (Chen et al., 1985; Hamadani et al., 2011; Hawkesworth et al., 2013; Ng et al., 2003; Tsai et al., 1999; WHO, 2011). In the Bengal Delta, elevated As concentrations occur primarily in Holocene sediments deposited less than 12 kyr ago, whereas As concentrations in pre-Holocene sediments are consistently <10 μg/l in most areas (BGS and DPHE, 2001; Burgess et al., 2010; McArthur et al., 2004; McArthur et al., 2008; Ravenscroft et al., 2005).

Lowering exposure by targeting deep (>150 m), low-As groundwater in pre-Holocene aquifers for installation of new wells has become a key form of mitigation and is likely to remain so for the foreseeable future

(Ahmed et al., 2006; DPHE & JICA, 2010a, 2010ab; Ravenscroft et al., 2013). Currently, several hundred thousand deep (>150 m) tube wells, hereafter referred to as DTWs, typically hand-pumped, are used for mitigation (DPHE & JICA, 2010a, 2010ab; Ravenscroft et al., 2014). Groundwater flow and As transport modeling predict that pumping low-As water from deep aquifers should be sustainable for hundreds of years or longer if high rates of concentrated deep pumping for municipal and irrigation supply are avoided (Michael & Voss, 2008; Shamsudduha et al., 2019).

Several studies have reported elevated As concentrations (often >50 μg/L) in deep groundwater on both sides of the border between the Indian state of West Bengal and southwestern Bangladesh (McArthur et al., 2016; Mukherjee et al., 2011; Ravenscroft et al., 2014; UNICEF, 2011). The cross-border nature of the contamination probably hid its full extent, as national-level maps would only show a portion. This contamination is cause for concern because deep groundwater is not a viable mitigation option in these areas and, perhaps more importantly, it might be an early sign that continued reliance on deep, low-As groundwater is not a sustainable solution. Human development could be the cause of arsenic contamination if deep mechanized pumping, which has been reported in the affected region of West Bengal (McArthur et al., 2016; Mukherjee et al., 2011), has already induced downward flow of As contamination from shallower aquifers. Understanding the origin of this deep, high-As concentrations therefore has important implications for As mitigation throughout the Bengal Basin and other As-affected regions.

In principle, pre-Holocene aquifers that are naturally low in As could become contaminated as a result of direct *transport* or *local release* of As caused by dissolved organic carbon (DOC) present in the sediment or transported from elsewhere. DOC triggers in situ reduction of Fe oxides and release of As to groundwater (Mailloux et al., 2013; McArthur et al., 2004; McArthur et al., 2016). Arsenic contamination resulting from transport has been attributed to municipal or irrigation pumping (Mukherjee et al., 2011; van Geen et al., 2013; Winkel et al., 2011) but could potentially also occur in areas with natural flow systems that transport Holocene-age groundwater to older sedimentary strata (e.g., Hoque et al., 2017; Mukherjee et al., 2011). Depressurization of deep mud layers by large-scale pumping could also release DOC or As locally without any inflow from a shallow contaminated aquifer (Chakraborti et al., 2009; Erban et al., 2014; Mukherjee et al., 2011; Planer-Friedrich et al., 2012; Winkel et al., 2011). The occasional contamination of a deep well due to the improper installation of a shallow screen or to flow through a broken well casing has also been documented, though the impact of such failures is likely to be limited (Choudhury et al., 2016; Lapworth et al., 2018).

We use inspection of well integrity, groundwater and sediment dating, and numerical groundwater flow modeling to determine the origin of unexpectedly high As concentrations in deep aquifers of the southwestern Bengal basin. We consider: (a) wells that are shallower than reported or broken; (b) rapid movement of Holocene groundwater to Pleistocene aquifers over the past 50 years due to pumping through tritium (^{3}H) groundwater age dating; (c) naturally occurring deep arsenic due to deeper than typical Holocene sediment using stratigraphic information and sediment radiocarbon (^{14}C) dates; and (d) natural movement of Holocene groundwater to Pleistocene aquifers over thousands of years using radiocarbon age of groundwater dissolved inorganic carbon (DIC) and groundwater flow modeling.

2. Methods

2.1. Field Survey and Data Collection

The study area is in the Ganges Delta in the SW Bengal Basin (Figure 1). Sampling was concentrated along two latitudinal transects across the India-Bangladesh border in winter 2015 and fall 2016. Samples were taken from village community supply wells claimed to be deep that are typically pumped at rates of approximately 1 m^3 per day. Initially, As was measured with field kits in 323 wells selected at regular intervals along the two transects (Arsenic Econo-Quick™) after purging each well for 5 to 10 min.

A subset of 55 tested wells showing high As concentrations based on the kit was selected for further investigation (Figure 1). Most of these wells were concentrated along the 100-km cross-border transect centered on the 23.2° N parallel, with an additional five wells located up to 30 km to the south (Figure 1). A downhole camera (GeoVISION Nano) was used to determine screen depths and check for obvious leaks or disconnected pipe sections.

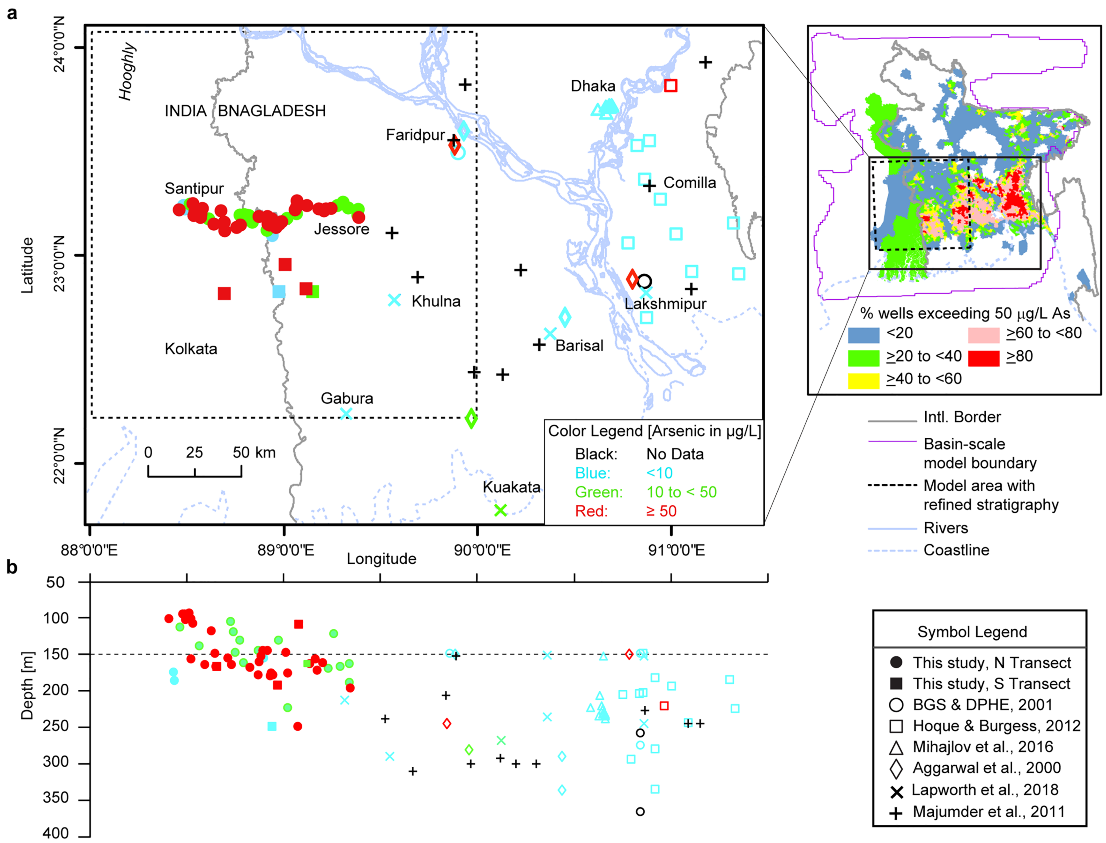

Figure 1. Map of locations of groundwater samples in the Bengal Basin, the basin-scale model area of Michael and Voss (2008), and the model area with refined regional stratigraphy. (a) Spatial distribution of deep wells with As and DIC ^{14}C age data, (b) depth distribution of the same wells. Groundwater As contamination dataset shown in the inset map of (a), dominated by field kit tests of shallow tube wells, are from NAMIC (2006) and Chakraborti et al. (2009).

After video verification of depth and screen length (Table S1), 33 wells with confirmed depth of 150 m were identified and sampled for DIC and ICPMS analysis for As. Nineteen were sampled for ^{3}H. Each well was pumped to remove three well volumes before sampling. DIC samples were collected in 250 mL glass bottles and poisoned with 1 mL of saturated $HgCl_2$. Four fossil wood and grass samples were collected for AMS radiocarbon dating from drill cores at two locations (Figure S4 and Table S2).

2.2. Groundwater Dating

The uncorrected and corrected radiocarbon ages were calculated considering the half-life of ^{14}C as 5,370 years and initial carbon activity of 100% and 90%, respectively. No other corrections were applied; further details are provided in Supporting Text. Water samples were collected in 500 mL glass bottles for ^{3}H and analyzed by mass spectrometry using the ^{3}He ingrowth method (Mihajlov et al., 2016) with a detection limit of ~0.01 TU.

2.3. Determination of Subsurface Lithological Variation

A total of 1,435 driller logs from a 200 × 180-km area surrounding the sampling transects were used to infer the regional (~100 km scale) hydrostratigraphy (Figure 3 and S3). Ten parallel lithologic panel diagrams were constructed in the east-west direction, each 20 km apart (Figure S3). The top and bottom boundaries of the main aquifer units were interpreted based on lithologic continuity. An aquifer unit was identified if the lithology was predominantly sand, and an aquitard unit was identified if the lithology was predominantly silt and clay. Areas with alternating sands and muds were left undifferentiated. Each line was digitized and three-dimensional maps of the top and bottom of the major hydrostratigraphic units were prepared by interpolation.

2.4. Determination of the Depth to the Holocene Sediments

Four wood samples from sediments at two locations within the study area were dated using radiocarbon method. Sediment ages determined by either radiocarbon or OSL methods at five other locations were selected from literature (Biswas et al., 2014; McArthur et al., 2008; Stanley & Hait, 2000). These ages, combined with the Last Glacial Maximum Palaeosol (LGMP) used as a stratigraphic marker where present (Hoque et al., 2012; Hoque et al., 2014; McArthur et al., 2008) were used to constrain the depth of Holocene sediments (<12 kyr) in the study area (Figure S4).

2.5. Groundwater Flow Modeling

The MODFLOW (Harbaugh, 2005) model of Michael and Voss (2008, 2009a, 2009b) for the Bengal Basin was used, with modification of aquifer properties within the study area for some runs (Figure S7). Slight modifications were made to the original digital elevation model to correct for vegetation in the Sundarbans and anomalous peaks in the eastern hills. Model sides are no-flow boundaries along basement rocks in the west, the Himalayas and the Shillong Plateau in the north, the Chittagong fold belt in the East, and >30 km offshore in the Bay of Bengal in the south (Figures 1 and S7). The top boundary was a specified head at land surface elevation representing the average shallow water table throughout the year in most areas. More details of boundaries, parameters, and calibration are given in Michael and Voss (2009a, 2009b). The flow simulation in this work was steady-state and did not include pumping, because the objective was to simulate age of groundwater in deep wells, which likely recharged well before any anthropogenic influence.

In much of the Bengal Basin, the aquifer system consists of alternating high- and low-hydraulic conductivity (K) materials with little regional lateral continuity. Representation as an equivalent homogeneous anisotropic medium is therefore appropriate for simulating the basin-scale average flow system (Michael & Voss, 2008, 2009b). The values of horizontal and vertical K ($K_h = 5 \times 10^{-4}$ m/s, $K_v = 5 \times 10^{-8}$ m/s) of the basin-scale model were retained outside of the study area. Within the study area, the K_v for the main aquifer unit delineated from stratigraphic analysis (Figures 3, S3, S7b), and both the K_h and K_v for the overlying surficial aquitard unit were varied (Figure S8). Pump test data (BWDB, 2013; Deshmukh et al., 1973; JICA, 2002) at 17 locations throughout the study area (Figure 3) give a K_h of ~$5 \times 10^{-5} - 5.05 \times 10^{-3}$ m/s (Table S3). A base-case value of $K_h = 5 \times 10^{-4}$ m/s for the main aquifer unit was therefore used. Because the SBA has a much lower proportion of mud and clay than other areas of the basin, K_v was assigned a value of 1×10^{-6} m/s, 100× higher than in the rest of the basin. A hydraulic conductivity of 1×10^{-7} m/s, typical for very fine sands and silts, was used as both the K_h and K_v of the surficial aquitard.

Sensitivity analyses were carried out that varied hydraulic conductivities and porosity to determine impacts on groundwater ages in the deep aquifers. Ages were compared to radiocarbon dates from the wells. Parameter values used in sensitivity analyses are given in Table S4 and results are presented in Figure S8.

The simulated travel time of groundwater recharge to depth was determined by advective particle tracking using MODPATH (Pollock, 2012). Each model cell that corresponded to the field location with a measured ^{14}C age was assigned 64 equally distributed particles tracked backward to recharge locations. The average of the 64 travel times is the simulated groundwater age for each location. Similarly, a map of groundwater age at a constant depth of 150 m throughout the study area was determined. An effective porosity value of 0.2 (Michael & Voss, 2008) was used.

3. Results

3.1. Well-Depth Verification and As in Groundwater

The depths of the 55 wells with As >50 μg/L according to the kit were verified with a downhole camera (Figure S1). The depths of 21 of these wells were misreported and 1 had a collapsed casing (Figure S1d). This left 33 (18/37 in India and 15/18 in Bangladesh) wells deeper than 150 m with confirmed screens at the bottom with an average length of 5.6 m and a maximum of 12.2 m (Table S1). The camera showed no clear indication of shallow leaks through disconnected pipes or multiple screens for any of the tested wells. Laboratory analysis confirmed the field results: 29 of the 33 wells had As concentrations above the WHO guideline of 10 μg/L and 22 had As concentrations of 50–140 μg/L (Figures 1 and 2a, Table S1). These results confirm that a broad swath of the Bengal Basin in the cross-border region has elevated As in deep groundwater.

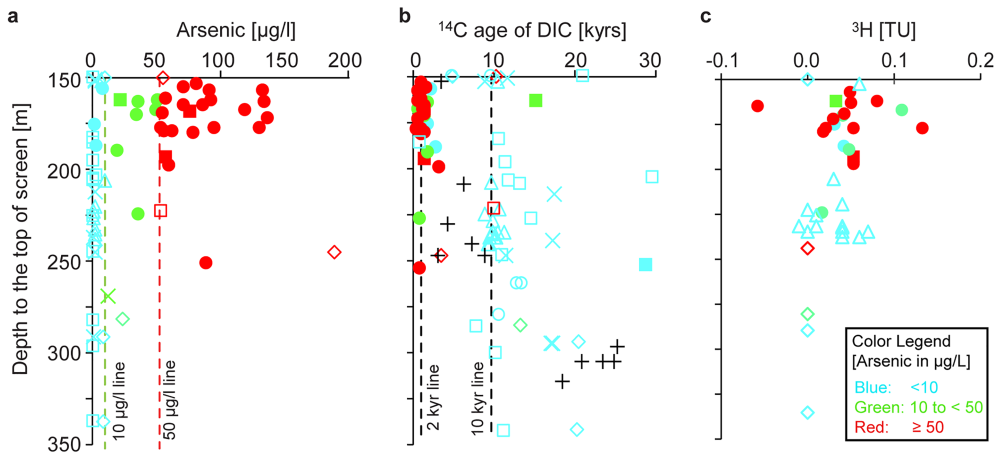

Figure 2. Depth distribution below 150 m of (a) As, (b) uncorrected ^{14}C groundwater DIC age, and (c) ^{3}H in deep wells from the Bengal Basin. Locations of all samples and legends are given in Figure 1.

3.2. Constraints on Groundwater Ages

Uncorrected radiocarbon ages in deep wells from the northern transect, irrespective of As concentration, were all <4.0 ^{14}C kyr, averaging 2.0 ± 0.6 ^{14}C kyr ($n = 29$, Figure 2b). There was no systematic trend with location or depth along the northern transect. In the two southern wells with high As, ages were also young, 2.0 ^{14}C kyr, but groundwater was much older in the other two low-As wells at 15.0 and 30.0 ^{14}C kyr

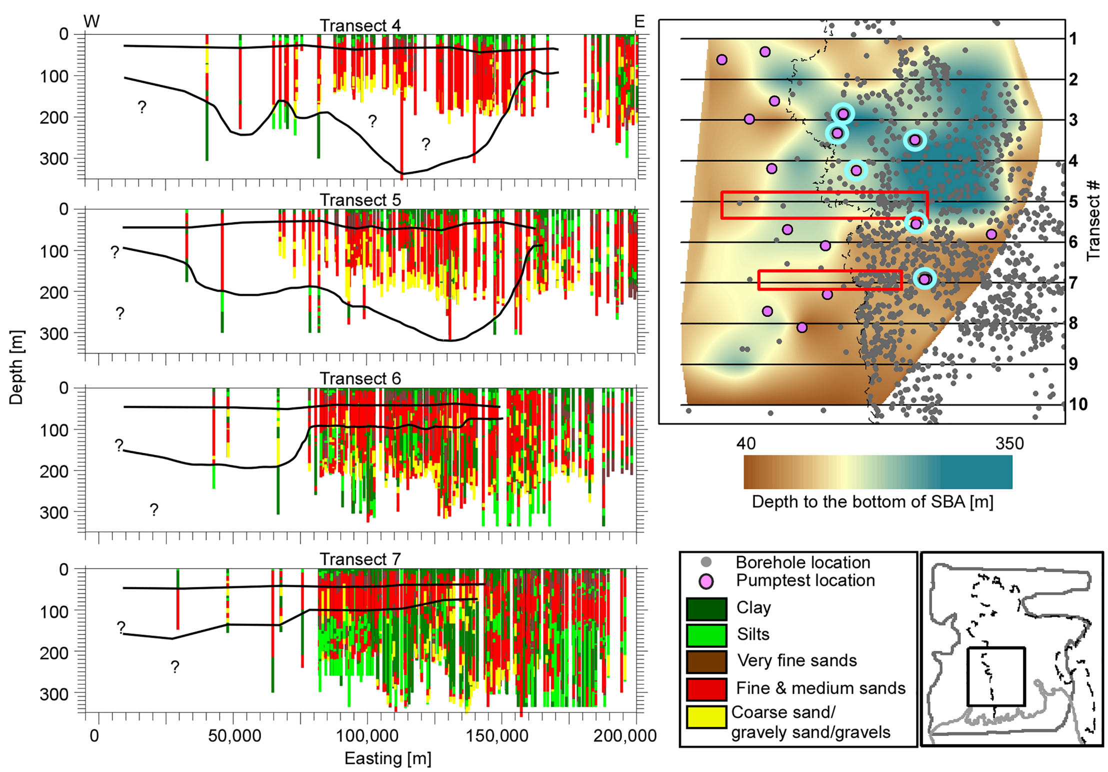

Figure 3. Delineation of regional hydrostratigraphy based on driller logs. Borehole locations are shown on the map. The lithologic panels are examples corresponding to line numbers shown on the map and display driller logs within 20 km swath of the line (see Figure S3 for all sections). The black lines on each panel represent the inferred top and bottom boundaries of the Sonar Bangla Aquifer (SBA). The colors on the map indicate the depth of the bottom of the SBA produced by digitizing and interpolating the lines drawn on the panels. The red rectangles outline the areas of sampled wells. The six coring locations of the Japan International Cooperation Agency (JICA, 2002) are marked with a cyan circle.

(Figure 2b). Corrections for the initial ^{14}C content of groundwater would reduce uncorrected ages by about 1.0 ^{14}C kyr, but with considerable uncertainty (Figure S2; Supporting Text S1).

Tritium measurements were used to identify water recharged in the past ~50 years. Concentrations of ^{3}H in most of the 19 tested wells >150 m clustered around 0.03 to 0.05 TU, except 6 with 0.05–0.13 TU (Figure 2c). The presence of measurable tritium indicates that at least some deep groundwater is young in age. We suggest that this result is most consistent with old groundwater mixing with a very small fraction of young groundwater containing bomb-tritium from testing of nuclear weapons in the 1950s (Craig & Lal, 1961; Kaufman & Libby, 1954; Lal & Peters, 1962; Munnich et al., 1966). Such a small contribution of young groundwater, perhaps caused by small leaks through the well annulus due to pumping (Lapworth et al., 2018), would not have significantly impacted radiocarbon in a larger pool of DIC (Mihajlov et al., 2016).

3.3. Stratigraphy and Age of Aquifer Sediments

Overall, the Bengal Basin aquifer system is highly heterogeneous, with a spatially variable distribution of fine-grained silt and clay units within a more sandy stratigraphy (Transect 6 in Figure 3 is typical for the Basin). However, the extensive set of driller logs reveals the presence of a sand and gravelly sand aquifer unit of variable thickness with little or no interbedded silts and clays within the study area at 20–300 m depth (Figure 3). This unit is termed the Sonar Bangla Aquifer (SBA) in Mukherjee et al. (2007). This aquifer is overlain by a 20 to 70-m thick surficial low permeability unit composed mostly of silts and very fine sands (the surficial aquitard). The depth to the bottom of the aquifer is more than 200 m near the northern transect, and it thins towards the south except along an NE-SW trending narrow channel-shaped area along which it is more than 100 m deep (map in Figure 3). Both the aquifer and the overlying aquitard gradually transition laterally into an alternating sand and mud sequence with little lateral continuity in individual sand and mud layers (Figures 3 and S3). Data indicate that the SBA sediments deeper than 20 to 70 m in the study area are likely to be >12 kyr old (Figure S4).

3.4. Simulated Age of Deep Groundwater

The homogeneous and anisotropic groundwater model (Michael & Voss, 2008) was calibrated with groundwater ages based on radiocarbon and head data available at the time of its development in 2008. Simulated ages with this model were greater than measured ^{14}C DIC ages for all samples in the northern transect and two of four in the southern transect (Figures 4a, 4c; Figure S5). This is the only region in the western part of the basin where the model systematically over-predicts travel time relative to radiocarbon ages (see Figure S5 for exceptions in other areas).

Updating the model by incorporating the regional hydrostratigraphy (Figures 3, S3, S7) virtually eliminated the mismatch between simulated and measured ^{14}C age in the study area (Figures 4b, 4c, 4e; Figure S5b). Similar improvement of model-simulated ages through incorporation of hydrostratigraphy was achieved by Shamsudduha et al., (2018) in the southeastern part of the basin. Sensitivity analysis indicates that this relationship remains true for a range of hydraulic conductivity values of the aquifer (Figure S8 and Supporting Text S1). In simulations with regional stratigraphy, recharge areas shifted 10's to 100's of km (Figure S6) and resulted in simulated travel times very close to the uncorrected ^{14}C ages for all but a few samples (Figures 4b, 4c, and 4 e). The oldest samples that show the greatest deviation from simulated ages (Figure 4b) are located in isolated sand lenses surrounded by clays (transect 7 in Figure 3). These secondary hydrostratigraphic features were not incorporated in the model.

4. Discussion and Conclusion

4.1. Origin of High-As Deep Groundwater

The origin of anomalous, relatively young, high As, deep groundwater in the southwestern Bengal Basin has significant implications for predicting current and future conditions in this and similar As-affected basins. We examine here the possible sources, argue for a natural, anomalous, deep groundwater flow system in the affected area, and discuss the implications of this finding.

Both physical and geochemical evidence indicate the elevated concentrations of As in deep groundwater of the study area are due to a natural deep flow system resulting from sand-dominated regional stratigraphy. Numerical modeling results provide a physical basis to show that groundwater in this geologically distinct

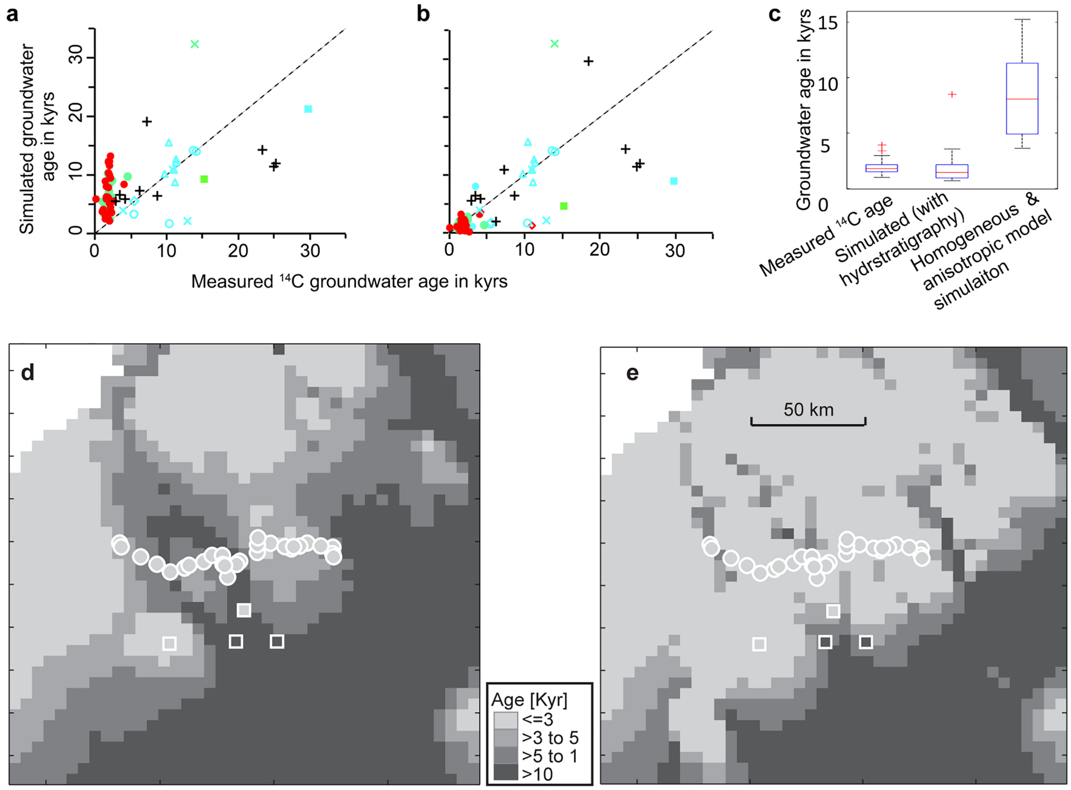

Figure 4. Comparison of measured ^{14}C age and simulated travel times. Scatter plots between uncorrected ^{14}C age and simulated travel time for all data west of the Meghna River (Figure 1) for (a) homogeneous-anisotropic model of Michael and Voss (2008) and (b) model incorporating regional hydrostratigraphy. Symbols as in Figures 1 and 2. (c) Measured and simulated groundwater ages for samples from the northern transect only. Boxes extend from the 25th to 75th percentile, red line is median, whiskers are range, outliers (data point beyond 1.5 times of interquartile range) are '+'. Simulated travel times to each model cell, 150 m below land surface for (d) Homogeneous-anisotropic model, and (e) model with regional hydrostratigraphy. Measured ^{14}C ages shown in symbols on same color scale and outlined in white. In white areas, the basement is shallower than 150 m, so travel time was not simulated.

area can have residence times shorter than those at similar depths in surrounding areas. The simulated residence times are consistent with deep groundwater radiocarbon ages that are all less than 4.0 ^{14}C kyr, indicating downward flow of Holocene-age, high-As groundwater to older strata deposited more than 12 kyr ago. This points to the anomalously high sand content of the study area sediments as a natural reason for high As at depth, given the contrast to other parts of the Bengal Basin where groundwater is both much older and low in As. The correspondence between simulated travel times and groundwater ages indicates a long-timescale process rather than recent fast transport from human perturbations. We note that the available data cannot distinguish direct input of As and input of DOC from above followed by local release of As at depth. The transport of both As and DOC is also retarded relative to groundwater flow by adsorption on aquifer sediments (Mailloux et al., 2013; Radloff et al., 2011), which further suggests that contamination of the deep aquifer predates human perturbation of the flow system (McArthur et al., 2016).

Despite this evidence, we consider whether pumping of deep groundwater, especially in areas where there are few clay layers above the pumping depth, could potentially have drawn Holocene-age high-As or high-DOC groundwater downward. Although large quantities of irrigation water are drawn from deep wells in many areas of West Bengal (McArthur et al., 2016; Mukherjee et al., 2011), this is not the case in Bangladesh where most irrigation wells extract shallow groundwater (<100 m). Irrigation pumping therefore cannot explain comparable levels of contamination on both sides of the border. Deep pumping for the municipal water supplies of Kolkata and Dhaka, with very large populations of 14 and 19 million, respectively, has caused a dramatic decline of 40–80 m in local groundwater heads that impacts deep aquifers over distances of up to 20–30 km (Khan et al., 2016; Knappett et al., 2016; Sahu et al., 2013). However, the study area is 80–180 km from these urban centers outside the area of influence. Deep pumping for smaller cities within the study area on either side of the border (Figure 1) including Santipur (pop. 0.29 million) and Jessore (pop. 0.25 million) is unlikely to be sufficient to affect the regional deep groundwater flow system, although contamination due to deep pumping on a very local scale is possible (Lapworth et al., 2018; Mukherjee et al., 2011). Most notable, however, is that none of the municipal pumping in these areas is deeper than 150 m. Therefore, deep penetration of high As concentrations in this part of the Bengal Basin is unlikely to have been caused by groundwater pumping (McArthur et al., 2016).

4.2. Implications for Deep Groundwater Vulnerability for As Transport

The results of this analysis show that transport of As into deeper, pre-Holocene strata can occur, with the potential to cause widespread contamination. While the timeframe over which this occurred in our study area is not well-constrained, modeling suggests that it required thousands of years. Contamination in our study area is therefore probably not a predictor of imminent widespread contamination of deep groundwater in other areas in the Bengal Basin where groundwater is significantly older. The stratigraphic anomaly that promotes naturally deep groundwater flow and contamination of deep groundwater with As is localized in the southwest of the Bengal Basin.

In heterogeneous depositional environments such as this fluvio-deltaic system, stratigraphic understanding provides clues to arsenic distributions. While small-scale heterogeneities have been shown to explain some of the As variability within Holocene aquifers (Aziz et al., 2008; Stute et al., 2007), larger-scale geologic features control groundwater flow, residence time, and associated As distributions, at increasingly deep, more regional scales. Thus, hydrostratigraphy may be used to target deep groundwater for testing prior to widespread investment for As mitigation in areas where water quality data is not available. Similarly, where lithologic data are sparse, consideration of depositional setting (e.g., Hoque et al., 2012) may help identify areas at risk for deep, high As concentrations. In the southwestern Bengal Basin, the sand-dominated lithology is attributable to a low subsidence rate leading to repeated lateral channel migration and the preservation of coarse over fine-grained channel sediments (Goodbred et al., 2003). Other areas where fine-grained sediments tend not to be preserved, for example beneath structurally constrained reaches of major rivers, may be similarly vulnerable.

This study highlights the importance of considering multiple factors—stratigraphy, hydrology, biogeochemistry, and human activities—that contribute to the spatial and temporal distribution of groundwater arsenic in large basins. These factors vary widely both regionally and locally. Thus, while large-scale models and geochemical analyses provide insights into management of the deep groundwater resource for sustainability, measurement of As concentrations after well installation and continued monitoring are essential to ensure the quality of drinking water in the Bengal Basin.

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Geophysical Research Letters

Supporting Information for

High-arsenic groundwater in the southwestern Bengal Basin caused by a geologically controlled deep flow system

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Text S1. **Supporting Methods**

^{14}C Age correction: Converting radiocarbon ages to calendar ages is notoriously difficult for DIC in groundwater because several poorly constrained corrections can subtract hundreds to a few thousand years [*Geyh,* 2000]. One method is to infer an initial radiocarbon age from the youngest sample in a data set that does not contain any detectable bomb-produced ^{3}H and therefore no bomb-produced ^{14}C either [*Hoque et al.,* 2012, *Mihajlov et al.,* 2016]. Using existing dataset, this would amount to a decrease in the uncorrected ages by about 1.0 ^{14}C kyr (Supporting Figure S2). Additional corrections reflecting the release of carbon along the flow path have been proposed [*Hoque et al.,* 2012, *Mihajlov et al.,* 2016] considered on the basis of the stable isotopic composition ($^{13}C/^{12}C$) of DIC but they are highly uncertain. The average $\delta^{13}C$ of -4.5‰ (σ = 2.2‰, n= 33) in deep groundwater DIC within the study area shows considerably less depletion than deep groundwater in other parts of the basin, with an average $\delta^{13}C$ of -16‰, (σ = 6‰, n= 39) (Figure 2c). We do not have sufficient data for the detailed geochemical modeling required for correction in such a case [*Aravena et al*., 1995] and no further corrections were attempted.

Sensitivity Analysis. The uncertainty in simulated ages was explored with sensitivity analysis. Groundwater age is highly sensitive to the vertical K (K_v) of both the SBA and the surficial aquitard unit as well as the horizontal K (K_h) of the SBA (Figure S3). Pump test data [*Deshmukh et al.,* 1973; *JICA,* 2002; *BWDB,* 2013] at 17 locations throughout the study area (inset map Figure S3) indicate that the hydraulic conductivity of the SBA ranges between approximately 5.22×10^{-5} and 5.05×10^{-3} m/s (Table S3), one order of magnitude variation from the base case value. Age is insensitive to the K_h of the surficial aquitard. Both the median and range of groundwater age in the northern transect increase with decreasing K_v, and both the median and range decrease with increasing K_v (Figure S8). However, the magnitude of the changes is larger for a decrease in K_v than an increase. An order of magnitude increase in K_v of the surficial aquitard compared to the base case reduces the median and standard deviation of simulated groundwater age by 70% and 16% of the base case values, respectively. In contrast, lowering the K_v of this unit by the same order of magnitude increases the median and standard deviation by 360% and 83%, respectively. Increasing the K_v of the SBA from the base case does not affect the median and standard deviation of simulated travel time (<2% change). However, a two order of magnitude decrease results in a 220% and 100% increase in the simulated median and standard deviation, respectively. Changes in K_h by an order of magnitude has a negligible effect on the simulated median age; however, the range of the simulated age changes proportionately.

Supporting Figures

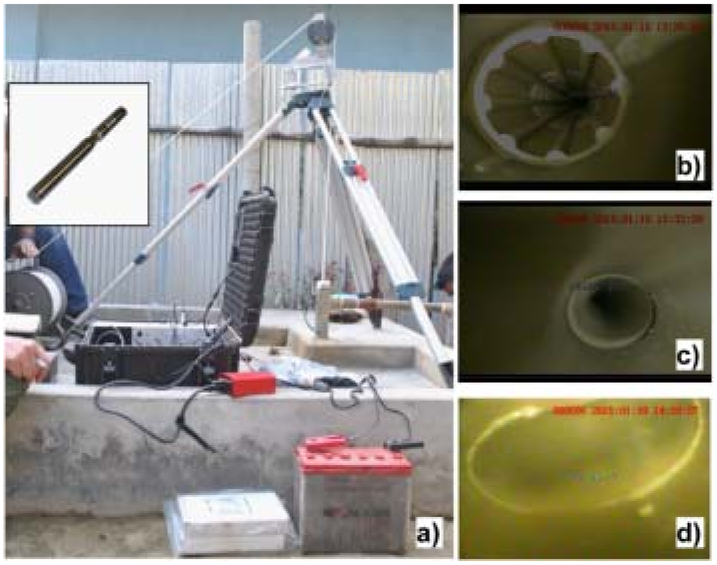

Figure S1. a) Field setup of downhole camera used in this study, camera is shown in the inset. Photos of b) typical well screen, c) joints in well casing, and d) broken well casing.

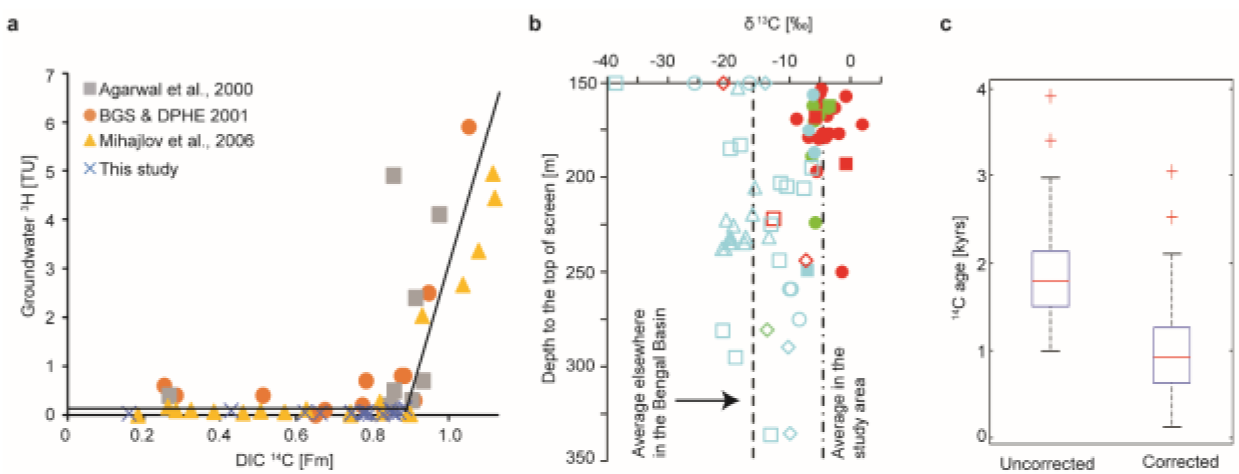

Figure S2. a. Empirical relationship between groundwater ^{3}H and ^{14}C in DIC plotted for all available samples from this study and other studies in the Bengal Basin. b. depth distribution of DIC $\delta^{13}C$. **c.** comparison of corrected and uncorrected ^{14}C ages of DIC in groundwater from the northern transect. Symbols in **b** are the same as in Figure 1.

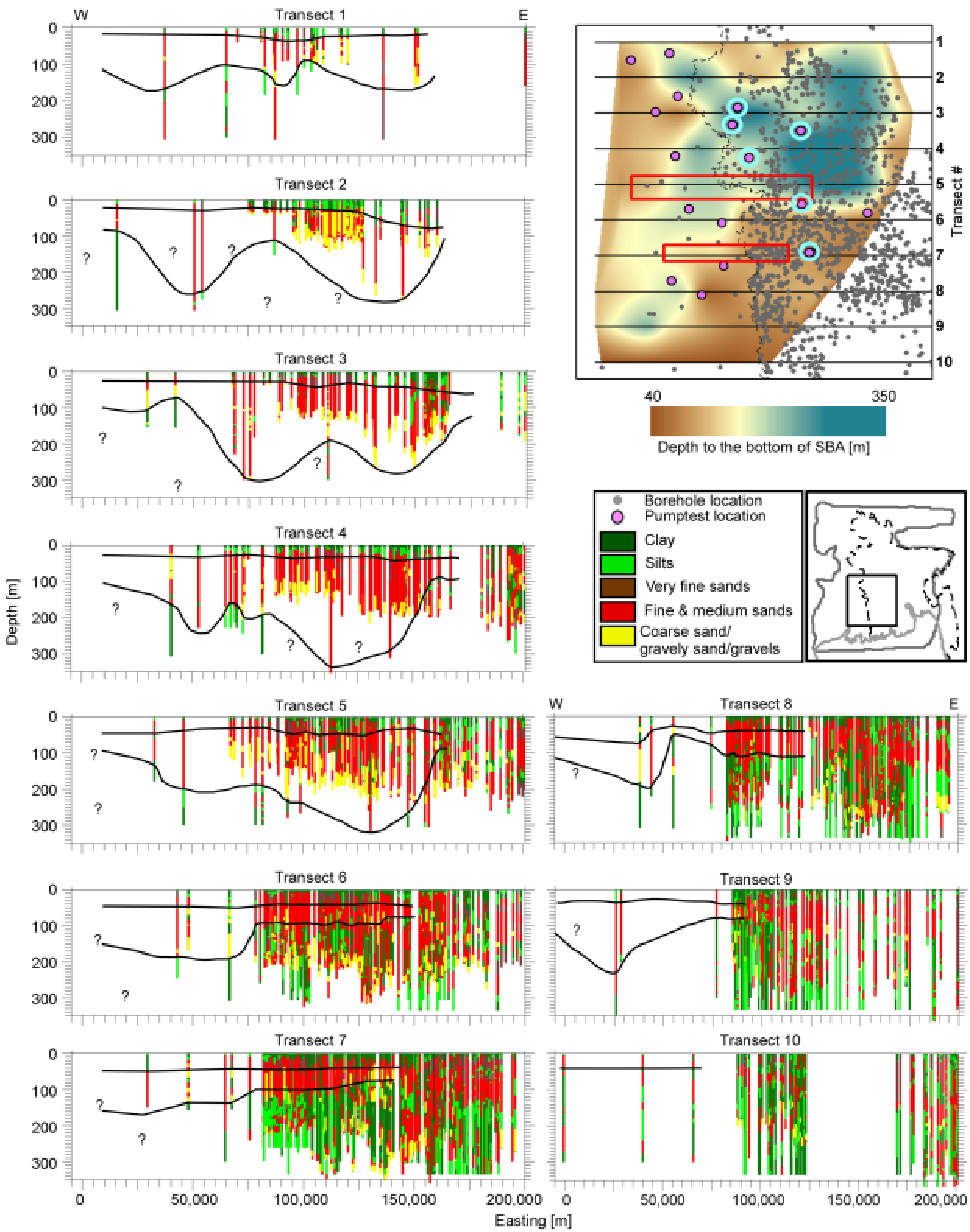

Figure S3. Delineation of regional hydrostratigraphy based on driller logs. Borehole locations are shown on the map. Each of the lithologic panels shown corresponds to lines # shown on the map and displays all driller logs within a 20 km swath centering on the line. The black lines on each panel represent the inferred top and bottom boundaries of the Sonar Bangla Aquifer (SBA). The colors on the map indicate the depth of the bottom of the SBA produced by digitizing and interpolating the lines drawn on the panels. The red rectangles outline the areas of sampled wells. The 6 coring locations of the Japan International Cooperation Agency [*JICA,* 2002] are marked with a cyan circle.

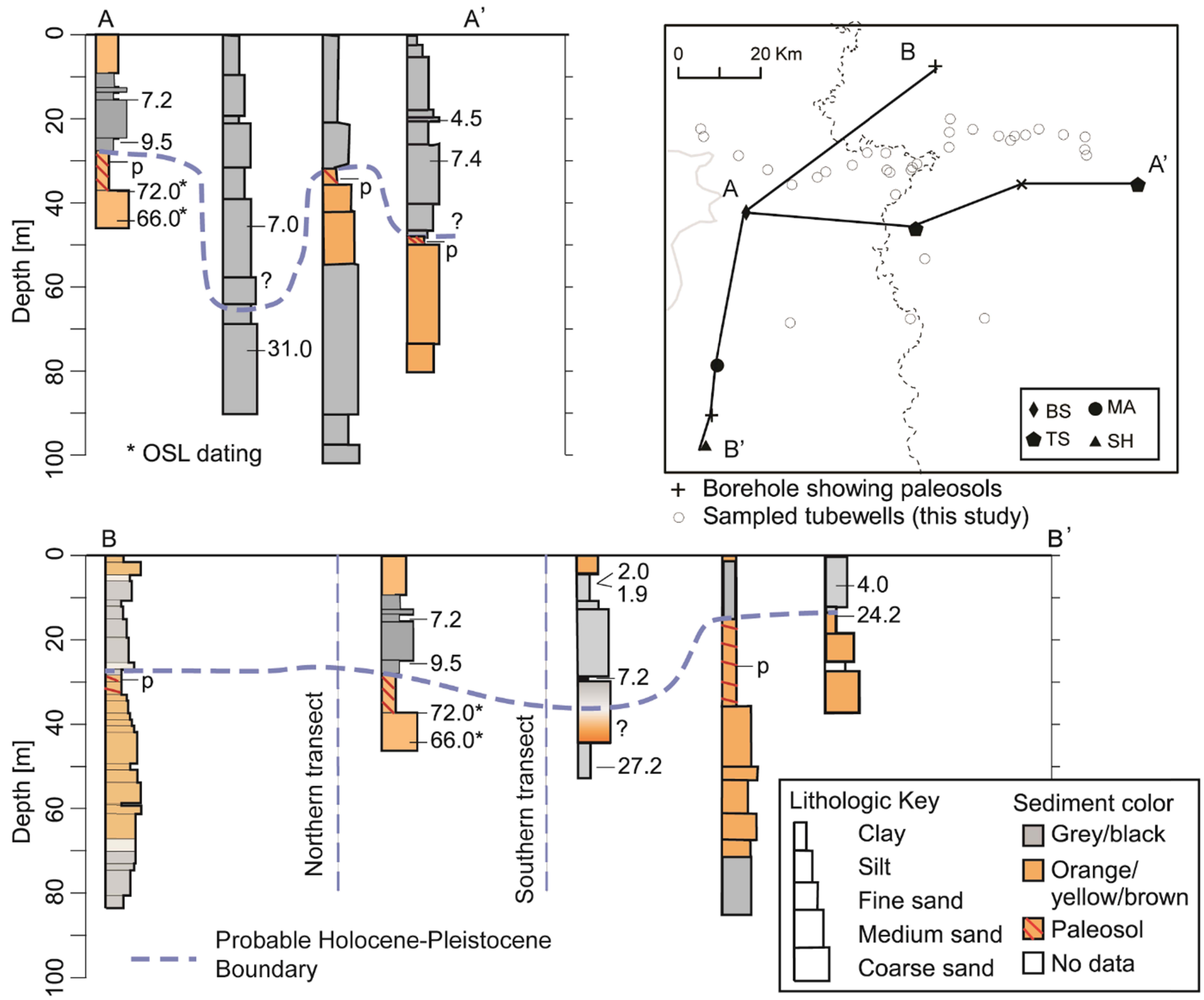

Figure S4. The shallowest reported Holocene-Pleistocene boundary in SW Bengal Basin based on sediment radiocarbon dates and the last glacial maximum paleosols (p). Ages are given in thousands of years at their respective positions on driller logs. Locations of transects and all available sediment age data in this region are shown in the inset map and on the lithologic panel BB', which runs approximately perpendicular to both transects (data for all locations shown on map are given in Supporting Table S3). Radiocarbon data sources: BS – *Biswas et al.* [2014]; MA - *McArthur et al.* [2008]; SH - *Stanley and Hait* [2000]; and TS – This study. Source of borehole logs showing paleosol only (+ symbols): *Hoque et al.* [2012; 2014] and (x symbol) *JICA* [2002].

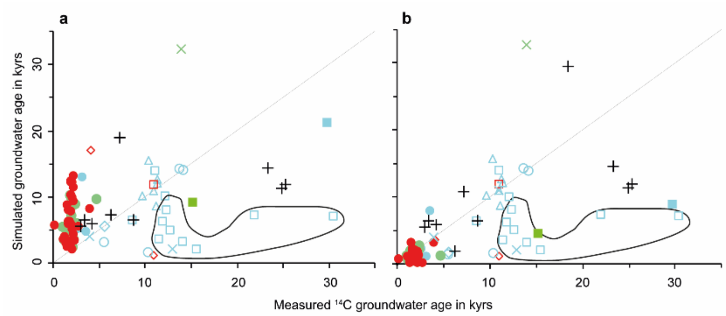

Figure S5. Comparison of measured ^{14}C ages and simulated particle travel times for all available data in the Bengal Basin (Figure 1). Scattered plots showing the correlation between the uncorrected ^{14}C age and simulated travel time for (a) homogeneous-anisotropic model of *Michael and Voss* [2008] and (b) model incorporating regional hydrostratigraphy. For symbols in both (a) and (b) see legend in Figure 1 or 2. The black polygons in both panel (a) and (b) outline the samples from east of the Meghna River (Figure 1). Simulated ages for locations east of the Meghna River are lower than measurements, likely due to regional stratigraphy (i.e., the eastern fold belt resulting in higher anisotropy than in the greater basin model); this does not substantially affect the simulated flowpaths in the study area. Indeed *Shamsudduha et al.,* [2019] found a good match between model simulation mand measured ^{14}C age in the SE part of the basin by incorporating hydrostratigraphy in their model. Three ^{14}C ages near Barisal shown in Figure 1 (two from Aggarwal et al. [2000] and one from Majumder et al. [2011]) are not shown in this plot. Wells are 290 to 335 m deep, with uncorrected ages from 20,777 to 21,303 years. Simulated ages are much greater, ranging from 61,860 to 112,192 years. The reason for this could be the evolution of hydrogeologic conditions in the basin over this time, hydrostratigraphic variation, or errors associated with sampling, such as misreported depths.

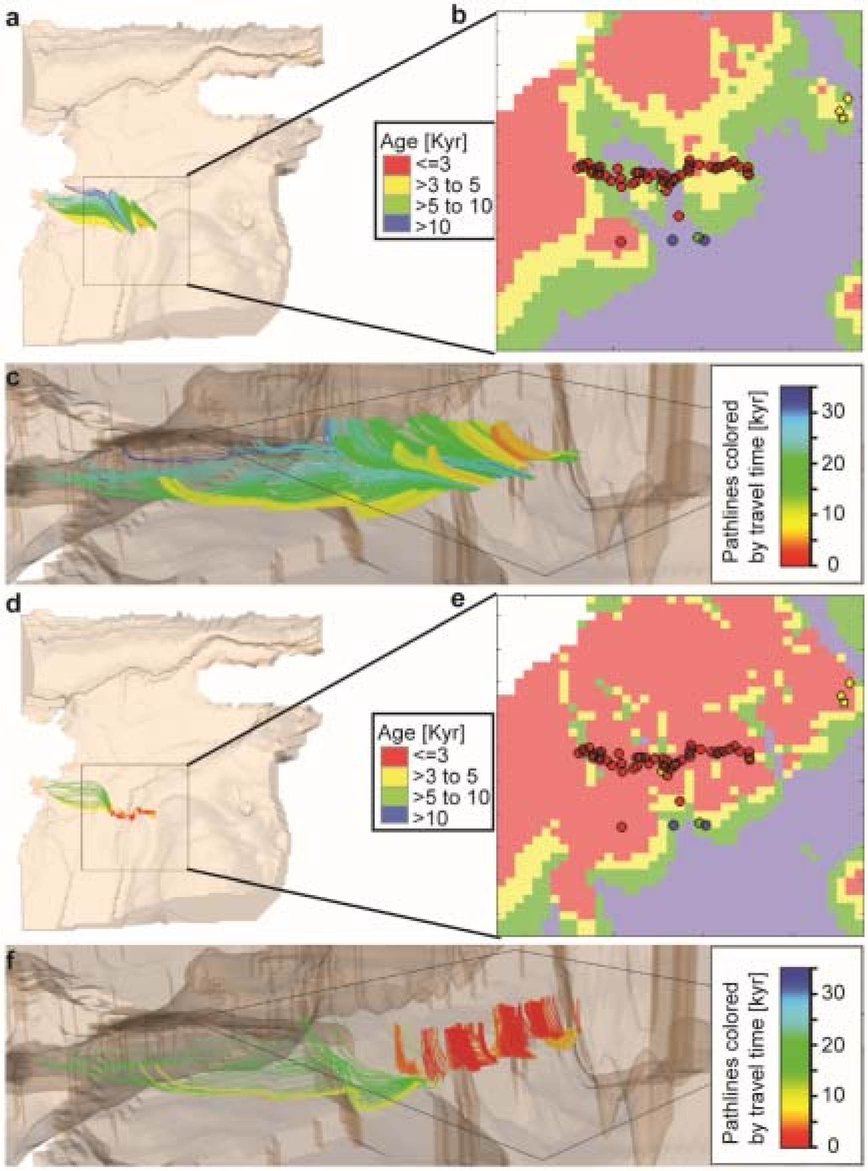

Figure S6. Simulated pathlines and travel times for two sets of aquifer properties. Panels **a**, **b**, & **c** are pathlines for all samples in the northern transect, simulated groundwater ages at a depth of 150 m below land surface, and zoomed in version of the pathlines shown in **a**, respectively for homogeneous and anisotropic aquifer properties. Simulated age is essentially the time a particle takes to travel from the land surface (recharge locations) to 150 m depth. The circles in panel **b** represent the measured ^{14}C ages. Within the white areas the basement is shallower than 150 m. Panels **d, e** & **f** are equivalent of panels **a**, **b**, & **c**, respectively but for the model scenario that incorporates the regional hydrostratigraphy.

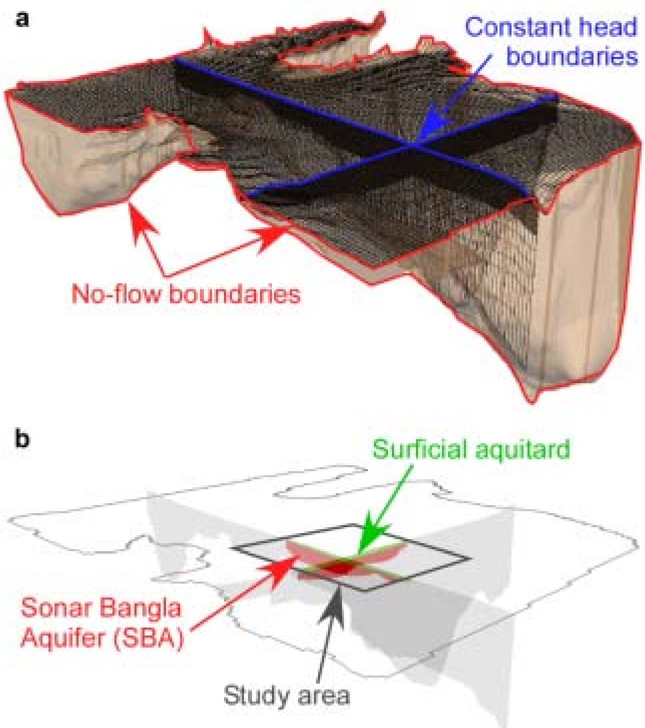

Figure S7. The Bengal Basin Model of *Michael and Voss* [2008, 2009a, 2009b]. (a) Model grid and boundary conditions. (b) The extent of the SBA and overlying aquitard represented in the model.

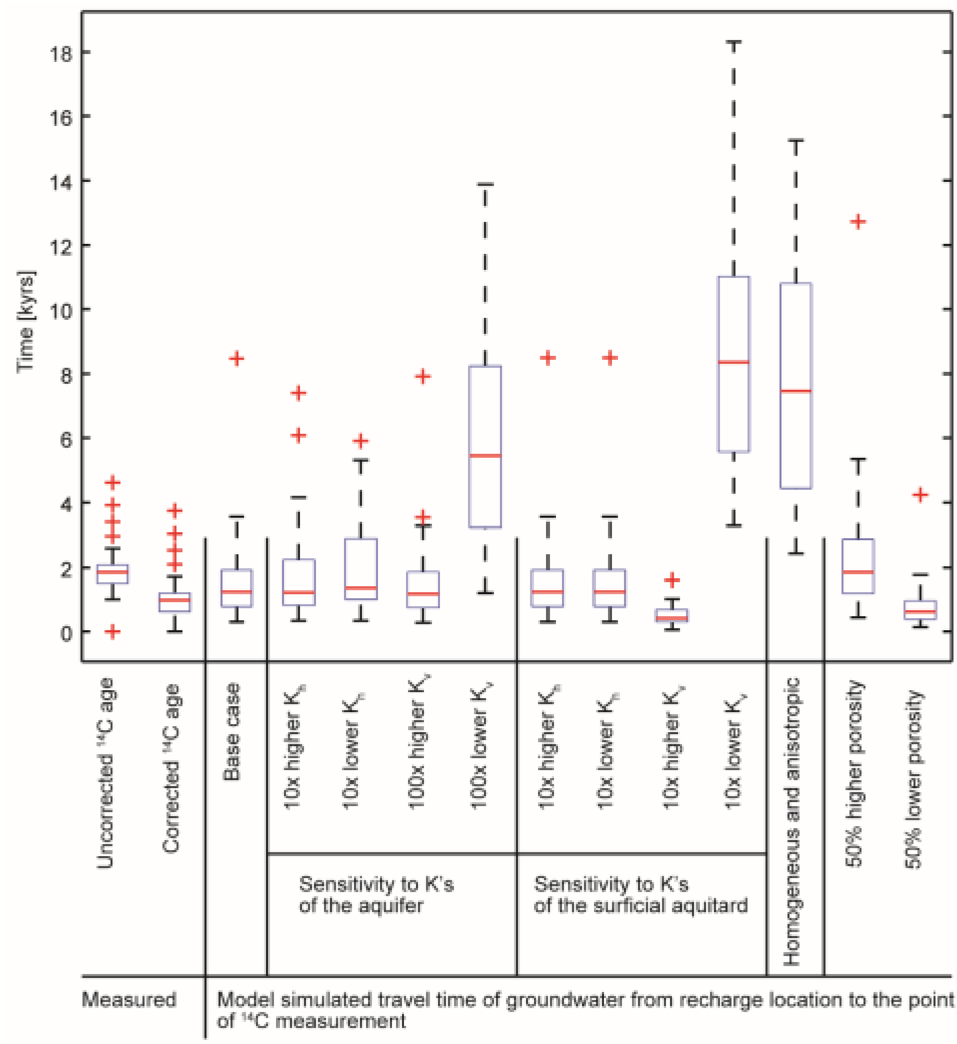

Figure S8. Sensitivity of simulated groundwater age to horizontal and vertical hydraulic conductivity and porosity. Each box extends from the 25th to 75th percentile, red line represents the median, whiskers show the range, and the outliers are shown by a '+' sign. Corrected ^{14}C ages were corrected only for initial ^{14}C activity.

Supporting Tables

Table S1. Groundwater arsenic, ^{14}C and $\delta^{13}C$ in DIC, and ^{3}H data of deep wells in the study area.

Well ID	Lat	Long	Depth [m]	Screen Length [m]	As [ppb]	DIC [mmol/kg]	FM ^{14}C	±σ [FM]	$\delta^{13}C$	Uncorr. ^{14}C age	Corr. ^{14}C age	^{3}H [TU]	±σ [^{3}H]
Northern Transect													
SAN40413	23.258	88.431	175		0.52	9.35	0.7443	0.0017	-6.84	2441	1570	0.0311	0.0143
SAN40411	23.241	88.439	187		1.75	9.35	0.6629	0.0016	-5.92	3399	2528	0.0418	0.0134
BAS41011	23.126	88.898	156	5.5	6.62	9.3	0.6978	0.0021	-6.08	2975	2104		
JOY40082	23.264	89.343	189	4.0	18.2	5.16	0.7466	0.0018	-6.34	2416	1545	0.0478	0.0138
LEB40003	23.279	89.233	170	4.0	33.5	9.68	0.7727	0.0019	-5.91	2132	1261	0.0407	0.0119
IND40033	23.236	89.343	163	4.0	34.9	7.6	0.7461	0.002	-3.53	2421	1550		
BEG40026	23.234	89.021	224	6.1	35.3	11.09	0.8273	0.0021	-5.74	1567	696	0.0167	0.0131
AZM40078	23.268	89.292	167	4.0	49	7.39	0.8469	0.002	-5.13	1374	503	0.1090	0.0180
BEA41003	23.188	88.793	162	7.3	49.9	10.1	0.8076	0.0018	-6.13	1766	896		
BAG40506	23.187	88.936	177		52.8	9.61	0.816	0.0023	-4.53	1681	810		
BAG40471	23.216	88.828	169		54.3	8.83	0.7791	0.0018	-8.82	2063	1193	0.0423	0.0107
BAG40459	23.217	88.872	179	5.5	55.4	7.67	0.7855	0.0016	-6.88	1996	1125		
SUN41007	23.179	88.876	162	6.1	56.3	9.22	0.808	0.0018	-5.6	1762	891		
BAG40029	23.223	89.347	197	7.9	58.9	7.81	0.6225	0.0022	-5.52	3918	3048	0.0530	0.0110
BAG40504	23.195	88.947	179	5.5	61.3	10.18	0.8341	0.0017	-4.08	1500	629		
GAD41001	23.159	88.711	155		70	7.71	0.7655	0.0014	-4.99	2209	1338		
KUJ41002	23.172	88.731	165	5.5	70.3	7.71	0.781	0.0014	-5.24	2043	1172		
BAG40502	23.181	88.933	180		78	8.68	0.8096	0.0017	-5.2	1746	875		
KUL41006	23.175	88.883	153	5.5	80.3	9.75	0.8096	0.0019	-4.75	1746	875		
RAN40538	23.173	88.593	165		85.2	9.63	0.7833	0.0018	-4.09	2019	1148		
MAR40024	23.274	89.076	250	9.1	87.4	7	0.8347	0.0019	-1.36	1494	623		
MAN40069	23.252	89.166	157	6.1	90.7	5.16	0.8519	0.002	-0.72	1325	454	0.04913	0.012
HAP40074	23.265	89.202	162	4.0	91.4	7.19	0.854	0.0021	-3.39	1305	434	0.08029	0.017
CHP40018	23.266	89.021	177	4.0	93.9	7.02	0.8732	0.0024	-1.89	1121	250		
MET41010	23.142	88.652	167		118	9.74	0.8019	0.0016	-3.82	1825	954		
NIA40025	23.295	89.022	177	4.0	129	5.17	0.8867	0.0021	-3.53	994	123	0.13269	0.014
ASH40567	23.202	88.523	157	5.5	133	9.22	0.7987	0.0018	-5.55	1858	987		
TIR40067	23.261	89.138	163	4.0	134	7.09	0.8187	0.002	-2.56	1654	783		
UTL40071	23.262	89.176	172	4.0	136	4.94	0.8576	0.0025	1.97	1270	399	0.02954	0.013
Southern Transect													
SWA40557	22.86	88.94	249	12.2	4.56	5.83	0.0277	0.0003	-7.07	29647	28776		
KHU40053	22.86	89.12	162	6.1	21.2	5.62	0.1606	0.0009	-3.48	15118	14247	0.03254	0.011
TEN40046	22.99	88.97	193	4.0	55.8	7.02	0.7846	0.0019	-0.72	2005	1134	0.05296	0.013
HAB40488	22.84	88.66	168		75.1	9.14	0.776	0.0017	-5.76	2096	1225		

Table S2. Sediment age data in and around the study area from literature

Area	Latitude	Longitude	Material	Depth [m]	Method	Age [ka]	AgeErr [ka]	Reference	Map ID
Barasat	22.742	88.488	Organic rich clay	7.6	^{14}C	2	0.1	[*McArthur et al.,* 2008]	MA
Barasat	22.742	88.488	Wood	7.6	^{14}C	1.9	0	"	"
Barasat	22.742	88.488	Organic rich clay	29.2-29.9	^{14}C	7.2	0	"	"
Barasat	22.742	88.488	Organic rich clay	49.5-50.4	^{14}C	27.2	0.4	"	"
Salt Lake (Kolkata)	22.567	88.467	Peat	4.25	^{14}C	4	0.1	[*Stanley and Hait,* 2000]	SH
Salt Lake (Kolkata)	22.567	88.467	Organic rich clay	13.2	^{14}C	24.2	0.6	"	"
Chakdaha	23.078	88.546	Peat	15.2	^{14}C	7.2	0	[*Biswas et al,* 2014}	BS
Chakdaha	23.078	88.546	Peat	25.8	^{14}C	9.5	0	"	"
Chakdaha	23.078	88.546	Quartz	37.3	^{14}C	72	7	"	"
Chakdaha	23.078	88.546	Quartz	43.6	^{14}C	66	7	"	"
Jessore	23.054	88.948	Wood	46	^{14}C	7	0.4	[*This Study*]	TS
Jessore	23.054	88.948	Wood	76	^{14}C	31.3	0.1	"	"
Narail	23.163	89.471	Wood	20	^{14}C	4.5	0.04	"	"
Narail	23.163	89.471	Wood	29	^{14}C	7.4	0.08	"	"

Table S3. Pump test data from the study area

Well ID	Latitude	Longitude	District	Locality	Reference	K [m/s]
WBBH08	23.85830	88.24170	Murshidabad	Takipur	[*Deshmukh et al.*, 1973]	1.10E-03
WBBH09	23.90000	88.45000	Murshidabad	Nawada		1.71E-03
WBBH11	23.38330	88.50000	Nadia	Bhatjangla		1.48E-03
WBBH12	23.68330	88.50310	Nadia	Debagram		1.02E-03
WBBH13	23.60000	88.38330	Nadia	Jugput		2.04E-03
WBBH14	23.11670	88.58330	Nadia	Neulia		1.43E-03
WBBH15	22.75000	88.50000	24—Parganas	Algaria		9.73E-04
WBBH16	22.68330	88.66670	24—Parganas	Berachampa		2.21E-03
WBBH17	23.05000	88.76670	24—Parganas	Khamarkulla		8.92E-04
WBBH18	22.83330	88.78330	24—Parganas	Dakshinchatra		1.12E-03
JICA-BH#1	23.63500	88.83420	Chuadanga	Chuadanga	[*Japan International Cooperation Agency*, 2002]	5.05E-03
JICA-BH#2	23.54920	88.80860	Chuadanga	Bara Dudpatila		1.75E-03
JICA-BH#3	23.52910	89.18190	Jhenaidaha	Jhenaidaha		1.41E-03
JICA-BH#4	23.38570	88.90460	Jhenaidaha	Krishna Chandrapur		1.19E-04
JICA-BH#5	23.15610	89.19860	Jessore	Jessore		8.10E-04
JICA-BH#6	22.91190	89.25670	Jessore	Rajnagar Bankabarsi		5.22E-05
JEKPOW2	22.91180	89.24540	Jessore	Keshabpur	[*BWDB*, 2013]	3.24E-04
NRNRPW	23.12000	89.56000	Narail	Narail		5.21E-04
					Minimum	5.22E-05
					Maximum	5.05E-03

Table S4. Hydraulic conductivity (K) values within the study area for each of the delineated hydrostratigraphic units for simulation scenarios. All K values are in ms^{-1}. Changes in parameter values in reference to the base case are highlighted.

Scenarios		**Surficial Aquitard**		**Sonar Bangla Aquifer**	
		K_h	K_v	K_h	K_v
	Base case	$1x10^{-7}$	$1x10^{-7}$	$5x10^{-4}$	$5x10^{-6}$
Sonar Bangla Aquifer	**10x higher Kh**	$1x10^{-7}$	$1x10^{-7}$	$5x10^{-3}$	$5x10^{-6}$
	10x lower Kh	$1x10^{-7}$	$1x10^{-7}$	$5x10^{-5}$	$5x10^{-6}$
	100x higher Kv	$1x10^{-7}$	$1x10^{-7}$	$5x10^{-4}$	$5x10^{-4}$
	100x lower Kv	$1x10^{-7}$	$1x10^{-7}$	$5x10^{-4}$	$5x10^{-8}$
Surficial Aquitard	**10x lower Kh**	$1x10^{-6}$	$1x10^{-7}$	$5x10^{-4}$	$5x10^{-6}$
	10x higher Kv	$1x10^{-8}$	$1x10^{-7}$	$5x10^{-4}$	$5x10^{-6}$
	10x lower Kv	$1x10^{-7}$	$1x10^{-6}$	$5x10^{-4}$	$5x10^{-6}$
	10x lower Kh	$1x10^{-7}$	$1x10^{-8}$	$5x10^{-4}$	$5x10^{-6}$
	Michael and Voss (2008)	$5x10^{-4}$	$5x10^{-8}$	$5x10^{-4}$	$5x10^{-8}$

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