

Vulnerability of low-arsenic aquifers to municipal pumping in Bangladesh



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SUMMARY

Sandy aquifers deposited >12,000 years ago, some as shallow as 30 m, have provided a reliable supply of low-arsenic (As) drinking water in rural Bangladesh. This study concerns the potential risk of contaminating these aquifers in areas surrounding the city of Dhaka where hydraulic heads in aquifers >150 m deep have dropped by 70 m in a few decades due to municipal pumping. Water levels measured continuously from 2012 to 2014 in 12 deep (>150 m), 3 intermediate (90–150 m) and 6 shallow (<90 m) community wells, 1 shallow private well, and 1 river piezometer show that the resulting drawdown cone extends 15–35 km east of Dhaka. Water levels in 4 low-As community wells within the 62–147 m depth range closest to Dhaka were inaccessible by suction for up to a third of the year. Lateral hydraulic gradients in the deep aquifer system ranged from 1.7×10^{-4} to 3.7×10^{-4} indicating flow towards Dhaka throughout 2012–2014. Vertical recharge on the edge of the drawdown cone was estimated at 0.21 ± 0.06 m/yr. The data suggest that continued municipal pumping in Dhaka could eventually contaminate some relatively shallow community wells.

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1. Introduction

Bangladesh has abundant, easily accessible groundwater within the unconsolidated, fluvio-deltaic sands that provide drinking water (Majumder et al., 2011) for 97% of its 160 million inhabitants (BBS, 2014). The most easily accessible shallow groundwater <100 m deep often contains toxic levels of arsenic (As) (DPHE/BGS, 2001), although its distribution is highly heterogeneous (van Geen et al., 2003). The As is mobilized from the surfaces of sediments in the Ganges–Brahmaputra–Meghna Delta (GBMD)

under reducing conditions associated with a supply organic matter (Harvey et al., 2002; McArthur et al., 2008; Neumann et al., 2010; Mailloux et al., 2013). Human exposure to toxic levels of As in drinking water has been documented to increase internal cancers and vascular diseases (Smith et al., 2000; Chen et al., 2011; Wu et al., 1989; Morales et al., 2000). In Bangladesh, villagers who had been consuming water with >150 µg/L As were shown to be twice as likely to die over a period of 8 years compared to villagers drinking water with <10 µg/L (Argos et al., 2010).

In some areas of Bangladesh, households can avoid exposure to As by switching to a neighbor's shallow low-As well (van Geen et al., 2002) but a more costly deeper low-As well is often preferred or required (van Geen et al., 2002; Chen et al., 2007). There are two types of deeper low-As wells: those privately installed in aquifers

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to depths of 30–90 m using the hand flapper drilling method and those installed using the donkey drilling technique in deep (>150 m) aquifer zones (Ali, 2003; Horneman et al., 2004). This latter approach is currently supported by the Bangladesh government (Ravenscroft et al., 2014) and as of 2011 the Department of Public Health and Engineering claimed they had installed 345,000 deep community wells throughout the country (Unpublished Data, DPHE), although other studies suggest this number may be somewhat lower (DPHE/JICA, 2010).

Nine million people live in the capital city, Dhaka. In 2011, an estimated 1.9×10^6 m³/day of groundwater was extracted by Dhaka Water Supply and Sewerage Authority (DWASA, 2012). Overall pumping for the greater Dhaka area is actually higher since another 5.5 million people live in the surrounding peri-urban areas (BBS, 2014). The annual volume pumped within Dhaka proper is equivalent to a 2.3 m-thick layer of water over the 300 km² area of this part of the city. As a result of a steady increase in pumping since 1983 the water table today lies 70 m below ground surface in some areas of the city (Fig. 1) (DWASA, 2012). The impact of massive urban pumping has been documented elsewhere (Ahmed et al., 1998; Hoque et al., 2007; Onodera et al., 2008; Barker and Lawrence, 2008; Hosono et al., 2009; Shamsudduha et al., 2009; Kagabu et al., 2011; Shao et al., 2013). Most studies have been concerned with water supply or quality immediately beneath a city, not in surrounding rural areas. Aquifer pollution concerns immediately beneath an urban landscape include oils, industrial solvents and fecal pollution from leaking sewer lines. The aforementioned studies have typically focused on the water quality and elevation of the water table immediately beneath a city rather than the potentiometric surface of deeper aquifers connected to a larger system of aquifers outside the city (Hoque et al., 2007). In cases where widespread geogenic contamination of shallow aquifers occurs the threat to deeper aquifers extends as far as the draw-down cone caused by urban pumping. The present study explores instead the consequences of the ongoing massive depressurization at depths >150 m below Dhaka on present and future access to low-As drinking water from aquifers tapped by wells located beyond the confines of the city (see Fig. 2).

Since millions of rural people rely on these low-As aquifers surrounding Dhaka for drinking water, urban pumping could increase human exposure to As from drinking shallow groundwater in two ways: (1) by rendering hand-pumps drawing water from the surface ineffective due to the depressurization of the deep aquifer system and (2) by inducing the downward migration of shallow, high-As groundwater into deeper low-As zones with the system (Michael and Voss, 2008). Migration of As into previously uncontaminated aquifers induced by municipal pumping has been postulated for the Indian portion of the Bengal basin and the Red River delta near Hanoi (Mukherjee et al., 2011; Winkel et al., 2011). Concentrated pumping could also indirectly cause the release of As to groundwater by downward flow of dissolved organic carbon (DOC) triggering the reductive dissolution of iron oxides (Harvey et al., 2002; McArthur et al., 2008; Neumann et al., 2010; Mailloux et al., 2013; van Geen et al., 2013). Finally, depressurization of the deep aquifer system could potentially release DOC and As from clay layers at depth, without any transport from shallow aquifers, as recently proposed for the lower Mekong delta (Planer-Friedrich et al., 2012; Erban et al., 2013).

In the present study, we evaluate the impact of urban pumping on hydraulic heads in the surrounding aquifer system located along a 35 km transect from Dhaka to the Meghna River on the basis of a total of 23 continuous pressure-logger records from 2012–14. Using this information, we then evaluate the impact of the regional depressurization of the aquifer system on access to low-As water and the longer-term risk of contaminating drinking water aquifers with As.

2. Study area and methodology

2.1. Site description

Dhaka lies near the confluence of three of the largest rivers in Asia: the Ganges, Brahmaputra and Meghna. The Ganges–Brahmaputra–Meghna (GBM) Delta is prone to flooding on a massive scale during the late monsoon (August–October) when the country receives the vast majority of its annual 2 m of rainfall. In extreme years as much as 2/3 of the country's land surface is flooded (Steckler et al., 2010).

The study area lies between central Dhaka and the Meghna River 30 km to the east, covering the eastern side of the Dhaka drawdown cone (Fig. 1). Within the study area lies a sub-district, Araihaazar, which is the primary geographic focus of this study. Araihaazar is the site of a 16-year running project to study the health impacts and origin of As exposure (van Geen et al., 2003; Argos et al., 2010). The sediment underlying most of Bangladesh contains unconsolidated sand, silt and clay. Three depth intervals are defined here for the purpose of this study: shallow (<90 m), intermediate (90–150 m), and deep (>150 m). Wells can be installed by a small team of local drillers using little equipment within 2–3 days to a depth of 90 m, whereas deeper wells require a heavier rig, more manpower, and up to a week (Ali, 2003). In Araihaazar, the distinction has no bearing on the As content of groundwater as low-As Pleistocene aquifers suitable for the installation of community wells that are both shallower and deeper than 90 m have been identified (van Geen et al., 2007; Dhar et al., 2008).

2.2. Spatial information

During a survey in 2012 the elevation of the tops of 110 well and piezometer casings in Araihaazar was obtained with a fast static GPS survey using two base station GPS receivers and one roving receiver (Trimble NetR9, Trimble Navigation Ltd., Sunnyvale, CA) (Steckler et al., 2010; Bauer-Gottwein et al., 2011). Post-processing of the 1-s measurements was performed with Trimble Business Center software (Trimble Navigation Ltd., Sunnyvale, CA). The exact positions (<1 cm error, xyz) of the base stations were determined using the software GAMIT (Herring et al., 2016) by post-processing with permanent base stations established throughout Bangladesh (Steckler et al., 2010). The GPS-derived elevations for the tops of well casings were challenged against a network of 9 wells whose relative elevations are known by repeat measurement with a water tube leveling survey and a theodolite (Marin et al., 2008). The total range of errors from this method, an estimate of precision but not accuracy, was ± 10 cm (Supporting Information). The elevation of the tops of two monitoring wells closer to Dhaka (U6 and BWDB) was estimated from the digital elevation model (DEM) built into Google Earth Pro (Google Inc.) with an error of ± 2 m (Haneberg, 2006).

2.3. Lithology

Well borehole lithology was obtained from geophysical logging and drill cuttings obtained from the two local drilling methods. With the reverse circulation (RC) hand-flapper method (Horneman et al., 2004), sediments are rapidly flushed up the inside of the Polyvinyl chloride (PVC) drilling pipe and this method appears to produce high quality lithologs (e.g. Zheng et al., 2005). With the forward circulation (FC) donkey drilling method, instead, water is flushed down the drilling pipe and travels up the annulus between the pipe and the sediments lining the borehole. The FC method therefore is more likely to mix sediments along the borehole wall. As a way of confirming cuttings-derived lithologies, a

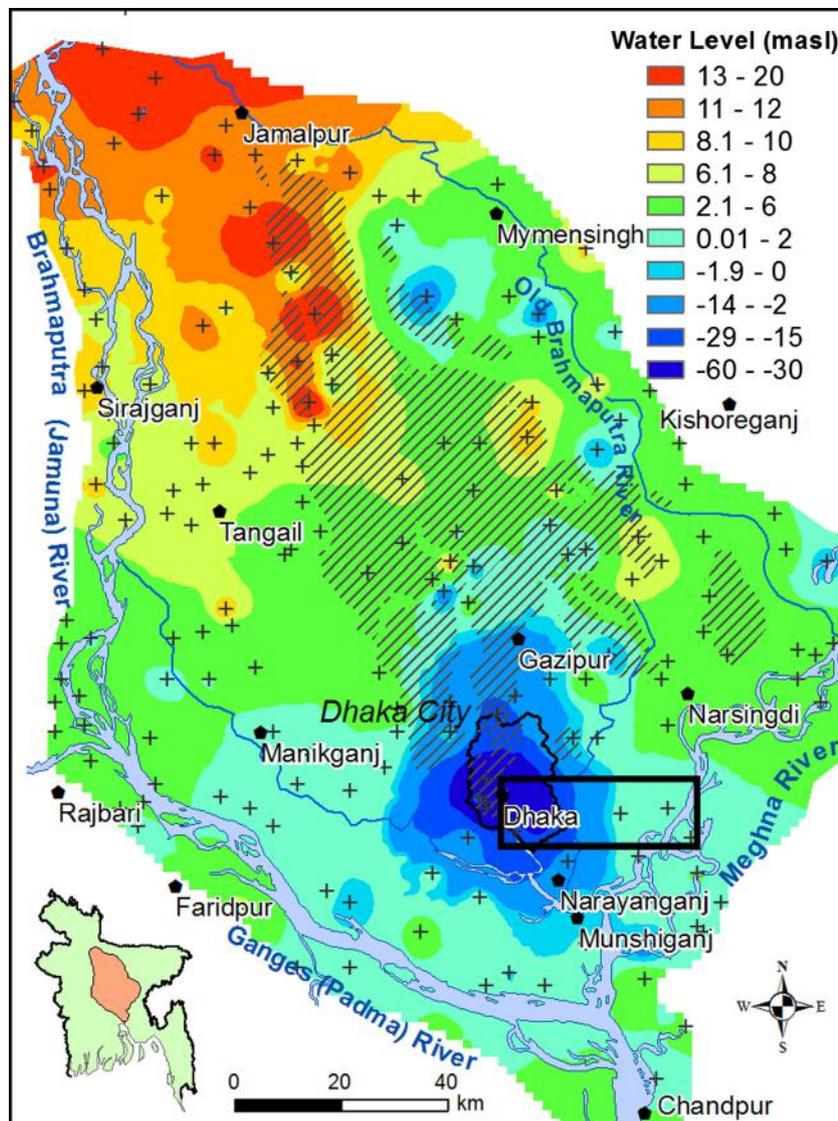


Fig. 1. Elevation of phreatic water table in central Bangladesh during the dry season in 2007. The black box encompasses the area discussed in this paper. Black crosses are locations of hydraulic head data. Diagonal hatched areas are Pleistocene uplands. See detailed explanation of the data sources for this figure in the Supporting information.

Geophysical logging sonde (1" Focused Induction Sonde/Natural Gamma, Robertson Geologing Inc., Houston, TX) was used to measure Electromagnetic (EM) conductivity and Natural Gamma Radiation of the sediment in a subset of six deep wells in eastern Araihaazar. A full list of all the wells used to obtain lithology information for this study are listed in the Supporting Information (Table S1). The sediments were broadly classified according to grain size and color¹ (Figs. 3 and S1–S3). Only three grain size classes were considered: silt or clay, fine-medium sand, and medium-coarse sand. Similarly two groupings of color were used: gray and brown, yellow or orange. The latter indicates oxidized sand, is often associated with Pleistocene age sediment, and contains low arsenic groundwater (Horneman et al., 2004; van Geen et al., 2007).

2.4. Well construction

Well construction determines how sensitive a well is to depressurization in the aquifer it pumps from. The elevations and construction details of 110 functioning community wells were

recorded. These had been installed throughout Araihaazar in 2001 and 2008 by the University of Dhaka and the non-governmental organization Water Aid, Bangladesh. The type of pump was recorded in 101 out of 110 cases. Eighty-one out of 101 pumps were above-ground suction pumps, referred to as "DTW6" in Ravenscroft et al. (2014). The other 20 wells had pumps that force water up from approximately 18 m depth using a plunger that runs along an inner PVC pipe inside the well. These are referred to herein as "Tara pumps". The 81 above-ground suction pumps are unable to pump water from deeper than approximately 9 m. Although the Tara pumps will continue to pump water when the water level is much lower than 9 m, they add about 10% to the ~USD1000 cost of installing a well with a DTW6 pump. Tara pumps are more difficult to repair because of limited supplies and expertise and thus they have a higher failure rate (Ravenscroft et al., 2014). The above-ground suction pumps are consequently more widely installed.

2.5. Water levels

Pressure transducers (Model 3001, Levellogger Edge, Solinst Canada Ltd., Georgetown, Ontario, Canada) were used to record

¹ For interpretation of color in Figs. 1 and 3, the reader is referred to the web version of this article.

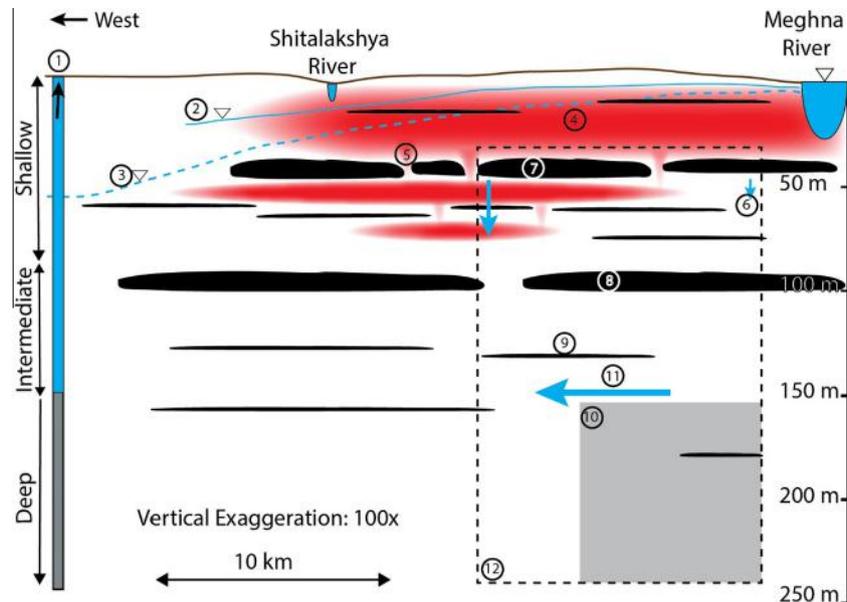


Fig. 2. Conceptual model of hydrogeochemical processes across the shallow, intermediate and deep aquifer systems in the outer Dhaka Drawdown Cone Area. (1) Municipal pumping wells in Dhaka; (2) unconfined water table outside of Dhaka; (3) potentiometric surface of the deep aquifer system; (4) shallow aquifers widely contaminated with geogenic As; (5) break in regional clay layers putatively allowing high As shallow groundwater to pass through; (6) observed vertical hydraulic gradients between the shallow and deep aquifer systems in central and eastern Araihaazar; (7) 15 m thick regional clay layer; (8) second 15 m thick clay layer widely present throughout eastern Araihaazar; (9) thin clay layers present throughout aquifer system; (10) intensively instrumented deep aquifer system; (11) flow direction in the deep aquifer system; (12) general study area and borders of Araihaazar.

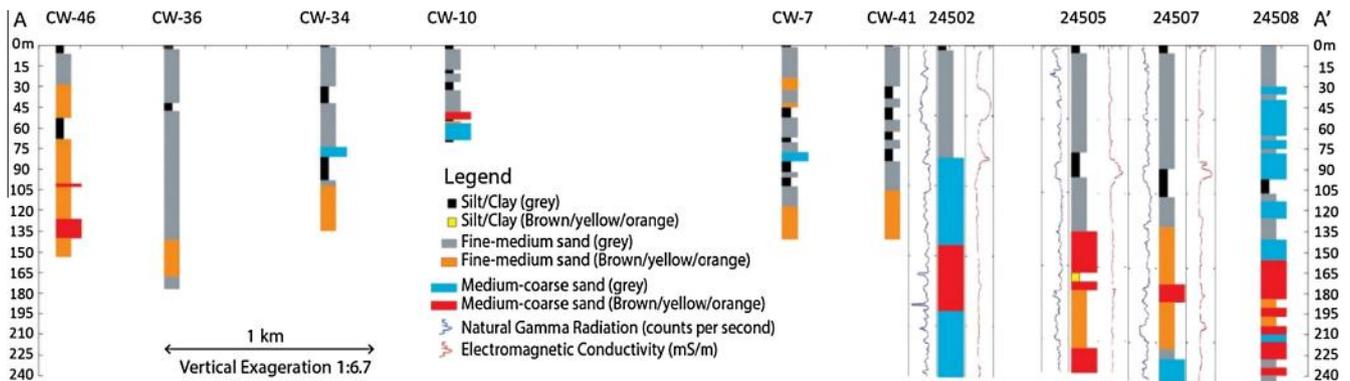


Fig. 3. Geologic cross section through Araihaazar A–A' (Fig. S1). All boreholes with ID's starting with "CW" (Community Well) were drilled using an RC method which produces excellent lithology. All other boreholes were drilled using an FC method which produces poor lithology. Electromagnetic (EM) Conductivity and Natural Gamma Radiation was recorded on some of the boreholes drilled with the FC method. Dual peaks in both the EM and Gamma logs indicate clay layers. For more information on the EM and Gamma methods see the SI.

water levels every 20 min. One logger was installed in a shallow private well in eastern Araihaazar; six were installed in shallow community wells; three were installed in intermediate wells; and twelve were installed in deep wells. One logger was installed in a river piezometer in the Meghna River on the eastern boundary of Araihaazar. Twenty-one of the 23 loggers had complete records extending from April 15, 2012 to June 1, 2014. A full list of the groundwater wells and river piezometer is provided in the Supporting Information (Table S2). The two deep monitoring wells closer to Dhaka were instrumented later. The pressure transducer records for well U6 and the Bangladesh Water Development Board (BWDB) well began on June 24, 2013 and July 7, 2014, respectively. The hydraulic head on July 7, 2014 from the BWDB well was used in the regional drawdown cone modeling (Section 3.1) whereas hydraulic heads from the rest of the wells were used from January 15, 2014.

Pressures were converted to water level elevation in two steps. The first step was removing atmospheric pressure fluctuations,

which were recorded by a Barologger (Barologger Edge, Solinst, Georgetown, Canada). The barometric pressure was measured in the center of the study area within 10 km distance of all the wells with pressure transducers. This step assumes a hydraulic efficiency of one implying no diminished effect of atmospheric pressure changes in the aquifers (PNNL, 1999; USGS, 2007). Water level fluctuations caused by community well usage prevented calculation of hydraulic efficiency. The day-time pumping disturbs the water levels so the first pressure measurement cannot be used for calibrating the transducers to a manually measured depth to water level. A second step was therefore followed to reference the median of the first 20 transducer pressure readings (first 6.67 h) to the manual water level measurement below the top of the well casing (Model 101, P7 Water Level Meter, Solinst Canada Ltd., Georgetown, Ontario, Canada). Each logger was calibrated twice; once at deployment and once after removal using the median of the preceding 20 pressures before removal. The same correction factor

was found in every case (± 1 cm) indicating that the method was effective and wire lengths did not change during deployment. A plane was fit to nighttime water levels in the deep aquifer every 24 h throughout the 2-year observation period using least-squares regression.

2.6. Groundwater arsenic

A blanket survey of Araihaazar was conducted in 2012–13 during which all 48,790 drinking water wells were tested for As using the ITS Arsenic Econo-Quick kit (<http://www.sensafe.com/481298.php>) and well owners were asked the depth and age of their wells (van Geen et al., 2014). The resolution of the test kit is limited compared with laboratory techniques with possible concentrations of 0, 10, 25, 50, 100, 200, 300, 500 and 1000 $\mu\text{g/L}$. In spite of this limitation, the kit correctly categorized As concentrations as being below or above the World Health Organization (WHO) and Bangladesh drinking water limits of 10 and 50 $\mu\text{g/L}$ most of the time (George et al., 2012; van Geen et al., 2014). A subset of 4811 wells of these wells was selected from the middle portion of Araihaazar spanning the distance from the western edge to the eastern edge at the Meghna River to visualize the typical spatial distribution of As for the present study.

3. Theory/Calculations

3.1. Regional drawdown cone modeling

To estimate the average Transmissivity (T) and Storativity (S) of the regional deep aquifer system observed water levels were modeled on the basis of the $1.9 \times 10^6 \text{ m}^3/\text{day}$ of groundwater reportedly extracted by DWASA in 2012 (DWASA, 2012). Two deep monitoring wells closer to Dhaka and 15 deep community wells in Araihaazar were used to constrain the regional shape of the drawdown cone (Table S2; Fig. 4). Several other deep community wells were included in the initial survey but not chosen for hosting a pressure transducer because they were too close to other wells at similar depth that already had one.

Four analytical models were tested to determine the best model to fit the observed shape of the regional Dhaka drawdown cone: (1) the confined Theis solution (Theis, 1935) (Fig. S4a); (2) the Theis unconfined solution wherein specific yield (S_y) is substituted for S (Freeze and Cherry, 1979) (Fig. S4b); (3) the Theis confined

solution assuming a fully penetrating constant head boundary along the Meghna River (Ferris et al., 1962) (Fig. S4c); and (4) the Hantush leaky aquitard solution (Hantush and Jacob, 1955) (Fig. S4d). Twenty years of steady pumping at the 2012 pumping rates was assumed. For the three confined models, T and S were varied systematically from 5.0×10^{-6} to 5.0×10^{-3} and 3.5×10^3 to $7 \times 10^5 \text{ m}^2/\text{day}$, respectively. Similarly, for the unconfined model S_y and T were varied from 0.1 to 0.5 and 3.5×10^3 to $7 \times 10^5 \text{ m}^2/\text{day}$, respectively. The leaky aquitard model (model 4) was chosen for use in this study since its optimal solution had the lowest Root Mean Squared Error (RMSE = 1.1 m) and Akaike's Information Criterion (AIC = 66) compared with observed heads. This ensures the most accurate and parsimonious model is chosen.

The Hantush leaky aquitard model (Hantush and Jacob, 1955) assumes the aquifer is confined, infinite in extent, isotropic and homogeneous with respect to T and S . The model also assumes radial, convergent flow towards the pumping center and predicts rapid stabilization of the potentiometric surface within approximately 100 days of a change in pumping rate (Fig. S5b). The model assumes that vertical leakage occurs according to Darcy's law across a continuous aquitard with a constant thickness (b') and hydraulic conductivity (K') proportionate to the difference in heads above and below the aquitard. These terms combine to make a single fitting parameter (b'/K') (Hantush and Jacob, 1955). Drawdown (s) within the aquifer at any distance (r) from the well at any time (t) is predicted by:

$$s(r, t) = \frac{Q}{4\pi T} W(u, \frac{r}{B}) = \frac{Q}{4\pi T} \int_u^\infty \frac{1}{t} e^{-\frac{r^2}{4B^2 t}} dt \quad (1)$$

$$u = \frac{r^2 S}{4Tt} \quad (2)$$

$$B = \sqrt{T \frac{b'}{K'}} \quad (3)$$

The term $W(u, r/B)$ (Eq. (1)) is the Hantush well function and r/B has been called "the dimensionless leakage parameter" (Neuman and Witherspoon, 1972). The solution for Eq. (1) was solved using a Matlab script provided in Veling and Maas (2010).

Modeled vertical recharge to the deep aquifer as a function of distance from the pumping center can be calculated using Darcy's law across the continuous aquitard using the local differences in hydraulic head between the deep and shallow aquifer (Fig. S5a).

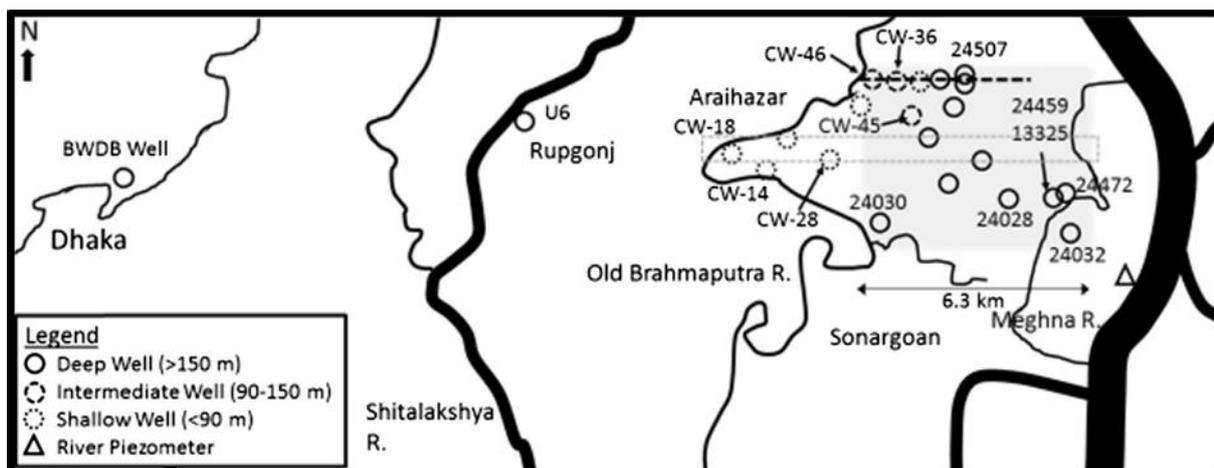


Fig. 4. Locations of wells and river piezometers used in this study. The shaded area represents the intensive $6.3 \times 6.3 \text{ km}^2$ study area. Water levels were monitored at 20 min intervals from June 1, 2012 to June 1, 2014. The dotted gray box indicates the area that As concentrations were reported in Fig. 8. Thick black lines represent major rivers, whereas thin lines represent inland streams.

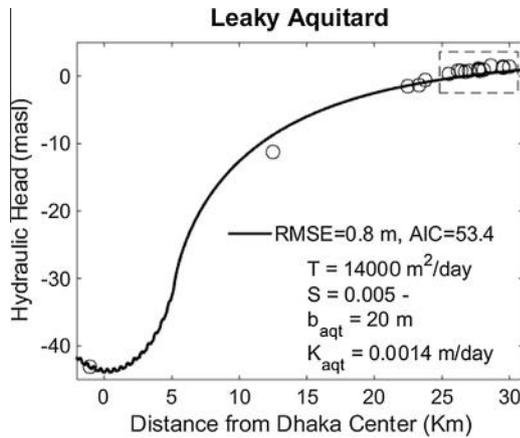


Fig. 5. Modeling Dhaka drawdown to estimate T and S of regional deep aquifer. The distance displayed is from 2 west to 31 km east of Dhaka center. The spikes at the bottom of each simulated drawdown cone is the result of combining 20 drawdown cones. The circles represent observed hydraulic heads. The dotted box demarcates the 6.3 km long detailed study area. Vertical exaggeration is 1:1000.

The optimal value of T was $1.4 \times 10^4 \text{ m}^2/\text{day}$. The optimal b'/K' ratio was 14,300 (day) (Fig. 5). We can assess how reasonable the fitted parameter values are by constraining the dimensions of the aquifer and aquitard, respectively. Transmissivity is defined as $T = K_h b$, where K_h is the average horizontal hydraulic conductivity of the aquifer system and b is its thickness. Therefore, the optimal T is equivalent, for example, to a deep aquifer system thickness of 200 m and horizontal hydraulic conductivity (K_h) of 60 m/day. The values for K_h in the deep aquifer agrees with past modeled (43 m/day in Michael and Voss, 2008) and measured values in shallow Bangladesh aquifers in Araihaazar (22 m/day in Nakaya et al., 2011; 35 m/day in Knappett et al., 2012).

The fact that the regional Hantush model fits the observations well does not guarantee it is an accurate conceptualization of the layered aquifer system. One of the assumptions of the Hantush model is that there exists an aquifer above the leaky confining unit whose hydraulic head remains constant across the pumping area. This is not accurate because the phreatic water table immediately within Dhaka is known to be dewatered in some places to 70 m depth (Fig. 1). The Hantush model reproduced the water levels accurately because a substantial amount of vertical recharge occurs across the pumping area proportionate to the amount of drawdown in the deep aquifer. The model indicates that 93% of extracted water from below Dhaka is vertically recharged within the drawdown cone area whereas only 7% comes from storage at depth. The optimal b'/K' ratio corresponds to possible (non-unique) values of b' and K' of 10 m and $7 \times 10^{-4} \text{ m/day}$, respectively. These are reasonable values based on available geologic data showing one or more 15 m thick clay layer between the surface and the deep aquifer system (Figs. 3, S2 and S3). Further, the pumping rate and its spatial distribution over greater Dhaka were not known accurately and the reported rate from DWASA was likely low. If the assumed pumping rate is increased 25%, this increases the value of T by 30%. This suggests that the values reported for T may vary by $\pm 25\%$.

3.2. Calculating local, daily vertical recharge into the deep aquifer system

In eastern Araihaazar, on the eastern edge of the Dhaka drawdown cone the shape of the potentiometric surface within the densely instrumented $6.3 \times 6.3 \text{ km}^2$ area of the deep aquifer system, combined with the estimated value of T (Section 3.1), permitted the calculation of vertical recharge at daily frequency using a

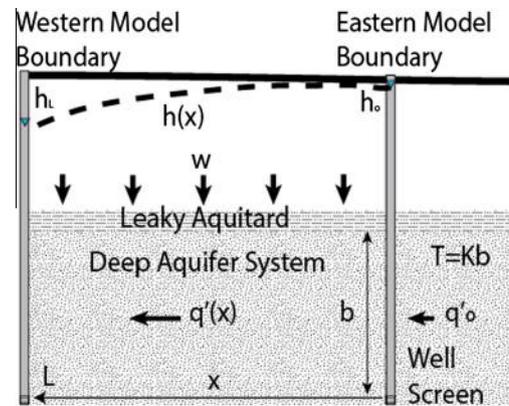


Fig. 6. Conceptual model used for the simulation of hydraulic heads and calculation of vertical and lateral fluxes to and from the deep aquifer system within the intensive $6.3 \times 6.3 \text{ km}^2$ study area in eastern Araihaazar.

1D local, analytical model with minimal assumptions (Fig. 6). This model assumes: (1) vertical recharge rate ($w \text{ [L/T]}$) and aquifer transmissivity ($T \text{ [L}^2/\text{T}]$) are uniform throughout the area; (2) flow within the aquifer system is horizontal; and (3) convergence in flow within the deep aquifer system at this distance from the center of pumping (23–29 km) is negligible and occurs along the east–west axis. This is an equilibrium model with respect to flow conditions as storage is negated. This is justified since fitted S values for the deep aquifer system (Section 3.1) applied to the observed hydraulic head fluctuations over the $6.3 \times 6.3 \text{ km}^2$ area indicated the annual total volume of water going into and out of storage ($6 \times 10^5 \text{ m}^3/\text{yr}$) was 1 order of magnitude lower than the annual flux across each of the model boundaries. Thus a steady-state assumption is reasonable for estimating fluxes across the model boundaries. The eastern and western boundaries of the model are defined by the hydraulic heads in the easternmost (24472) and westernmost (24030) deep wells, respectively (Figs. 4 and 6). We do not assume an explicit location for the upper or lower boundary of the modeled aquifer.

The lateral volumetric flux per unit width of the deep aquifer system ($q' \text{ [L}^2/\text{T}]$) at any point $x \text{ [L]}$ along the aquifer ($q'(x)$), is equal to T times hydraulic gradient at x . In this model x starts as zero on the eastern boundary and increases towards the west. At point x , flux is also equal to the lateral flux ($q'_0 \text{ [L}^2/\text{T}]$) on the eastern boundary of the model plus the cumulative vertical flux ($w x \text{ [L}^2/\text{T}]$) across the distance (x) from the eastern boundary ($x = 0$) (Eq. (4)).

$$q'(x) = -T \frac{dh}{dx} = q'_0 + wx \quad (4)$$

$$-T \int_0^x \frac{dh}{dx} dx = \int_0^x (q'_0 + wx) dx \quad (5)$$

$$T(h_0 - h(x)) = q'_0 x + \frac{w}{2} x^2 \quad (6)$$

$$h(x) = -\frac{w}{2T} x^2 - \frac{q'_0}{T} x + h_0 = -Ax^2 - Bx + C \quad (7)$$

$$w = 2TA \quad (8)$$

Both sides of Eq. (4) can be integrated from $x = 0$ to x to obtain a quadratic equation describing hydraulic head as a function of distance from the eastern boundary (Eq. (7)). Eq. (7) determines the shape of the potentiometric surface using the parameters: w , T , q'_0 and hydraulic head on the eastern boundary (h_0). The coefficients A , B and C (Eq. (7)) were optimized to the potentiometric surface using non-linear least-squares regression every 24 h. These

coefficients were then converted to physical parameters assuming a value for deep aquifer T estimated from modeling the regional Dhaka drawdown cone (Section 3.2). Specifically, the coefficient, A , in front of the quadratic parameter in Eq. (7) was used to determine the value of w (Eq. (8)).

The deep aquifer system can be modeled in 1D because the lateral hydraulic gradient is always due east. Although Dhaka pumping induces this gradient this flow model was chosen because the observed equipotential lines do not indicate converging flow paths. To confirm that the observed gradients could be created by Dhaka pumping without inducing convergent flow paths at this distance from the city forward modeling was performed with the calibrated Hantush radial flow model (Hantush and Jacob, 1955). The predicted equipotential lines in eastern Araihaazar produced by the Hantush model were straight (north to south), parallel and evenly spaced confirming negligible convergent flow. From a side view this surface has a linear hydraulic gradient confirming that any curvature in this surface results from local recharge.

4. Results

4.1. Lithology

We organized borehole cuttings into geologic cross-sections to describe the lithology of the aquifer system and provide context to the hydraulic calculations made in this study. The lithology of the aquifer system up to 250 m depth has not been previously published for Araihaazar at the ~15 km scale. Shallow lithology (<80 m) has only been published at a few sites at the village scale in western Araihaazar (Horneman et al., 2004; Zheng et al., 2005; Dhar et al., 2008). The lithologs obtained with the RC drilling method have more fine to medium sand than lithologs obtained with the FC method (Fig. 1). This indicates the RC method produces more accurate measurements. The color transition from reduced, gray sand, likely of Holocene age, to oxidized, orange/brown sand, likely of Pleistocene age, typically occurs between 60 and 100 m depth. The orange/brown sand is much shallower, however, in the north-west portion of Araihaazar where the transition occurs at 30 m in one borehole (Fig. 1). The oxidized sand is often capped by a thick sequence of clay from 30 to 50 m depth (Fig. S3). In addition to this clay layer, another thick layer is frequently recorded or inferred by EM/Gamma logs at 90–110 m depth in eastern Araihaazar where the wells were drilled deep enough to sample this depth (Figs. 3 and S1–S3). The RC drilled boreholes indicate many thin and discontinuous clay layers at shallow depths (<90 m) (Fig. 3). Fewer clay layers were recorded in the FC drilled boreholes in eastern Araihaazar, however, the EM/Gamma logs suggest that more may be present (e.g. well 24502 in Fig. 3).

4.2. Accessibility of low-As aquifers

Short-term variations in water levels indicative of pumping from the community wells ceased during the late dry season in 2 out of 9 wells in western Araihaazar in 2012–2013 (Fig. 7). In 2013–2014 this increased to 3 wells (Table S2). This phenomenon was observed whenever water levels dropped below 9 m from the surface (Fig. 8). For the most impacted well (CW-46), access to groundwater with a hand suction pump decreased from 72% of the year in 2012–2013 to 64% in 2013–2014.

The depth distribution and As concentrations for a subset of 4811 drinking water wells taken from the recent blanket survey of Araihaazar (van Geen et al., 2014) reveal how critical this problem is (Fig. 8). The portion of the aquifer system with the most abundant low-As wells (30–90 m) is included in the depressurized zone in western Araihaazar.

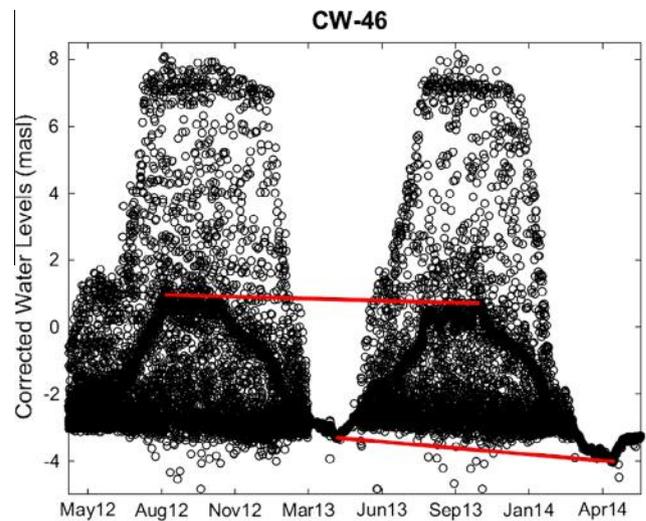


Fig. 7. Water Levels in a single community well on the western side of Araihaazar. Red lines represent the decreasing peak (-0.25 m/yr) and trough (-0.71 m/yr) water levels between 2012 and 2014. The absence of pumping noise indicates periods when water from well could not be accessed since it was too far below the surface for the suction pump to work. A moving 24 h trimmed mean where the upper and lower quartiles were trimmed, was used to calculate the peak or trough water level amidst pumping noise. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

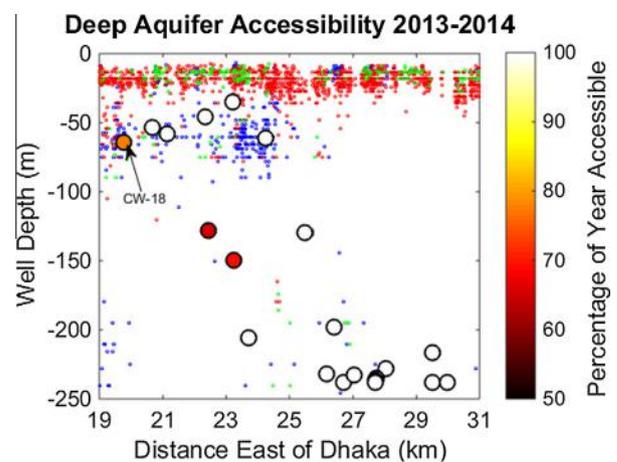


Fig. 8. Decreasing accessibility of low-As aquifers caused by Dhaka pumping. Large circles indicate screen locations of community wells equipped with pressure transducers. Water levels in three community wells were inaccessible for a substantial portion of the year in 2013–2014. The arrow indicates a community well that had accessible year-round water levels in 2012–2013. Small, colored circles represent the depth distribution and As concentrations in 4811 private and community drinking water wells that fell within a 1 km wide east–west swath through Araihaazar (Fig. 4). The arsenic data comes from an exhaustive 2012–2013 survey of 46,914 private and community wells across Araihaazar. Red, green and blue correspond to high (>50 ppb), medium (10–50 ppb) and low (<10 ppb) arsenic concentrations, respectively (van Geen et al., 2014). Vertical exaggeration is 1:48. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4.3. Inter-annual water level declines

Over the two years of monitoring, the water levels decreased for all 19 wells with data of sufficient quality to describe the troughs, while 17 of 19 wells with data describing the peaks showed decreasing water levels (Table S2, Fig. S6). In two cases each year, the pumping noise during a peak or trough was too extreme to obtain a reliable average water level for one day, hence records for 19 wells were retained instead of 21. The trough differences

varied from -0.17 to -0.74 m, while the peak differences varied from $+0.53$ to -0.82 m. The wells with the greatest declines were those screened below 50 m depth and closest to Dhaka (Fig. S6). Reported rainfall for Dhaka and three surrounding Bangladesh Meteorological Department (BMD) weather stations indicated stable rainfall amounts for the years from 2012 through 2014 (Fig. S7). This rules out decreasing recharge as a cause for the observed hydraulic head declines in the deep aquifer.

4.4. Seasonal variations in water levels

Each year the level of the Meghna River increased rapidly during the early monsoon, rising 3 m from its dry season level (Fig. 9a). It remained high until early October but by late November it rapidly dropped 3 m, where it remained low until the start of the next monsoon (May). Periodic tidal fluctuations are evident during the early monsoon and dry season when the level of the Meghna River was low and noticeably affected by semi-diurnal tides in the Bay of Bengal (Jakobsen et al., 2005; Steckler et al., 2010). A sine wave was fitted to these dry season oscillations using non-linear least-squares regression, confirming an approximately 14 day period corresponding to spring and neap tides (Steckler et al., 2010).

The hydraulic head at shallow depth (25 m) 2 km inland from the Meghna River was higher than the river throughout all seasons (Shallow Well East or 13325, Fig. 9a). The hydraulic head difference between the shallow well and the river peaked during the late monsoon and early dry season, and was close to zero during the late dry season when all hydraulic heads from the river and the adjacent aquifers converged (Fig. 9a). High frequency oscillations in the shallow water table caused by neighboring irrigation wells are visible during the late dry season, as previously reported elsewhere (Harvey et al., 2006).

In contrast, the hydraulic head in deep wells adjacent to the Meghna River (Deep Well East or 24459, Fig. 9a) lagged the sharp rise in hydraulic heads at shallow depths and the river during the early monsoon, rising 3 m in three months from April to July. Deep well hydraulic heads remained approximately 0.5 m below the level of the Meghna River until the end of the monsoon. During the early dry season the hydraulic head in the deep aquifer system decreased more slowly than the river resulting in a hydraulic head that was higher than the level of the river for most of the dry season.

On the western end of the study area, hydraulic heads in deep (Deep Well West or 24030) and intermediate (Intermediate Well West or CW-45) wells were lower than in deep wells in the east throughout the year (Fig. 9a). Hydraulic heads in shallow wells on the western boundary of the study area (Shallow Well West or CW-28) were much higher than in underlying intermediate and deep wells throughout the year (Fig. 9a).

4.5. Seasonal variations in lateral and vertical hydraulic gradients

The lateral hydraulic gradient in the deep aquifer system trended away from Dhaka throughout the year. The average lateral gradient for 2012–2013 and 2013–2014 increased from 2.7 to 2.8×10^{-4} , respectively. The analysis indicated the direction of steepest descent was consistently due West ($270 \pm 10^\circ$) (Fig. 10).

The magnitude of this eastward hydraulic gradient fluctuated seasonally and co-fluctuated with long- and short-term oscillations in the level of the Meghna River especially during the early monsoon (Fig. 9a and b) (Video S1 <https://youtu.be/951QEAvFZGk>). River levels and the lateral hydraulic gradient in the deep aquifer indicate four phases in the annual cycle: early and late monsoon (M1 and M2), and early and late dry seasons (D1 and D2). Throughout the two-year observation period, the lateral hydraulic gradient

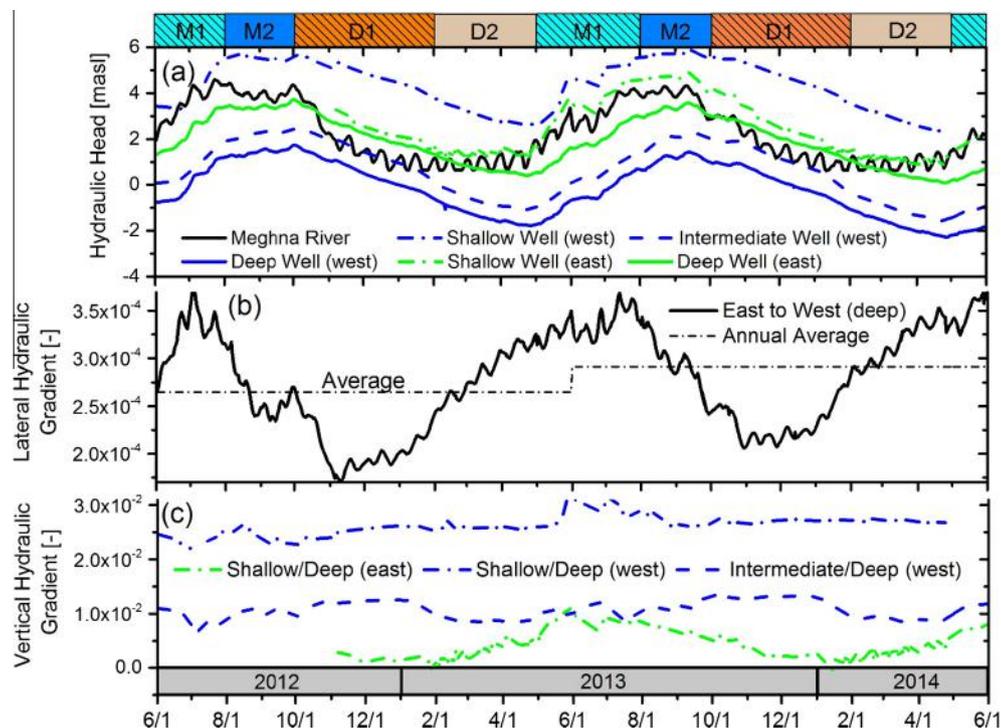


Fig. 9. (a) Seasonal relationships between hydraulic heads in wells emplaced within shallow and deep aquifers 2 km west of the Meghna River and 9 km west of the river. On the western end of the study area, Shallow, Intermediate, and Deep Wells correspond to wells CW-28, CW-45 and 24030, respectively. On the eastern end, Shallow and Deep Wells correspond to wells 13325 and 24459, respectively (Table S1). (b) Magnitude of average (linear) lateral hydraulic gradient within the deep aquifer. The horizontal dotted line represents the annual average lateral gradient. (c) Magnitude of the measured vertical hydraulic gradient between the shallow, intermediate and deep wells. Positive vertical hydraulic gradient corresponds to downward flow. M1, M2, D1 and D2 correspond to four seasons: early monsoon (ending Aug 1), late monsoon (Oct 1), early dry (Feb 1) and late dry seasons (May 1), respectively.

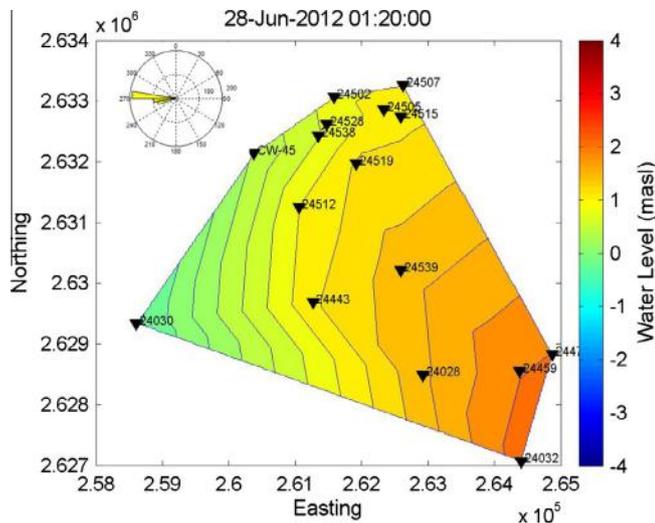


Fig. 10. Potentiometric surface of the deep aquifer in eastern Araihaazar during the early monsoon (June 28, 2012). The Rose diagram represents the average groundwater flow direction for 405 days at 1:20 am from April 15, 2012 to May 25, 2013.

ranged from a low of 1.7×10^{-4} in the early dry seasons to a high of 3.7×10^{-4} in the early monsoon. During the late monsoon the lateral hydraulic gradient rapidly decreased while the river level remained high. The early dry season was characterized by a continued decrease in the lateral hydraulic gradient accompanied by a sharp decline in river level. During the late dry season, however, the lateral hydraulic gradient increased steadily. At this time, the hydraulic gradient co-varied with the sharp, regular tidal fluctuations in the river (Video S1).

Vertical hydraulic gradients in Araihaazar favored downward flow between the shallow and deep parts of the aquifer system throughout the two years (Fig. 9c). On the western end of the study area, vertical gradients were much higher between shallow and deep wells than on the eastern end (Fig. 9c). This is because the deep aquifer system in the west is closer to the center of urban pumping in Dhaka. The vertical gradient between the shallow and deep aquifers in the west varied little seasonally. The gradient between the intermediate and deep aquifers however, increased somewhat during the early dry season. The calculation of vertical hydraulic gradients in the west is hindered by the lack of co-located wells of different depths. This was not the case on the eastern boundary of the study area, where a 25 m (13325) and a 238 m well (24459) were located only 50 m from each other. Here the vertical gradient peaked in the early monsoon and approached zero in the middle of the dry season.

4.6. Modeling the local deep aquifer system

The local 1D model can be used to calculate instantaneous recharge and discharge to and from the modeled deep aquifer system. In the early dry season vertical recharge peaked at 1.8×10^7 m³/yr while discharge occurred at both the eastern (-1.1×10^7 m³/yr) and western (-0.7×10^7 m³/yr) ends of the modeled deep aquifer system (Fig. 11a). This vertical recharge corresponds to an instantaneous Darcy Flux of 0.42 m/yr (Fig. 11a). During the late dry season and the early monsoon the vertical recharge decreased to near zero while lateral recharge on the eastern boundary increased. During the late monsoon vertical recharge increased again while the eastern boundary reverted from a recharge to a discharge boundary.

The net annual downward recharge entering the modeled local deep aquifer system in 2012–2013 and 2013–2014 was estimated

to be $8.6(\pm 2.1) \times 10^6$ and $8.9(\pm 2.2) \times 10^6$ m³, respectively (Fig. S8). The ranges represent an estimated 25% uncertainty in T derived from fitting the model (Hantush and Jacob, 1955) to the regional drawdown cone. The fitted, average, local vertical Darcy fluxes to the deep aquifer system for 2012–2013 and 2013–2014 were $0.21(\pm 0.06)$ and $0.22(\pm 0.06)$ m/yr, respectively. Assuming a porosity of 0.4 (Knappett et al., 2012) these correspond to average linear groundwater velocities of $0.54(\pm 0.14)$ and $0.56(\pm 0.14)$ m/yr, respectively.

5. Discussion

5.1. Hydrostratigraphy

The thick clay layer frequently observed in this study throughout Araihaazar at 30–50 m depth will tend to isolate the upper shallow (<30 m) aquifer system from the lower (>50 m) in those areas where it is present. Similarly, the second relatively continuous clay layer frequently observed at 90–110 m depth would tend to isolate the shallow aquifer system (<90 m) from the intermediate. These and the other thinner, more discontinuous clay layers observed likely confer a high hydraulic anisotropy (K_h/K_v) on the basin-wide aquifer system (Michael and Voss, 2008, 2009a). In contrast, below 110 m depth the EM/Gamma logs and the borehole lithology of the deep wells in eastern Araihaazar suggest that no thick clay layers are present. The deep aquifer system (>150 m) may therefore be more isotropic than the shallower aquifer system. The observed pattern of a thick clay layer overlying oxidized sand has been interpreted elsewhere in the GBM Delta as a paleosol, formed during the last glacial maximum (McArthur et al., 2008).

5.2. Accessibility to low-As aquifers

The results presented in this paper show that even in a water-rich region of the world, concentrated groundwater pumping may lead to safe drinking water scarcity. Depressurization of the deep aquifer system from Dhaka pumping is limiting access of people in rural areas to low-As drinking water. This is a water sustainability problem partly caused by a limitation of rural pumping technology as above-ground suction pumps are widespread in Bangladesh. Limited access to dry season water levels in community wells with suction pumps has been reported previously (Ravenscroft et al., 2014). A country-wide survey of 57,025 deep community wells that were installed by the government between 2007 and 2012 found 81% of these wells were equipped with above-ground suction pumps and the remainder had Tara or “forcing” pumps. After 6 years of operation, 16% of the Tara pumps were broken whereas only 7% of the simpler above-ground suction pumps were broken. Of the remaining 93% of mechanically functioning suction wells, the water level in 13% of them exceeded the maximum pumping depth for some fraction of the year. This is similar to the 14% (3/21) of monitored community wells with suction pumps that were inaccessible for some of the year in the present study. Our observations show that urban pumping has already reduced access to low-As groundwater in peri-urban areas and may do so to a greater extent in the future.

5.3. Water levels and hydraulic gradients

The year-round eastward trend in the average (linear) lateral hydraulic gradient within the deep aquifer system is consistent with an expanding Dhaka drawdown cone. Modeling suggests that the Meghna River is not currently a major source of recharge to the affected deep aquifer system with a net recharge across the eastern boundary of the modeled 6.3×6.3 km² area on the outer fringe of

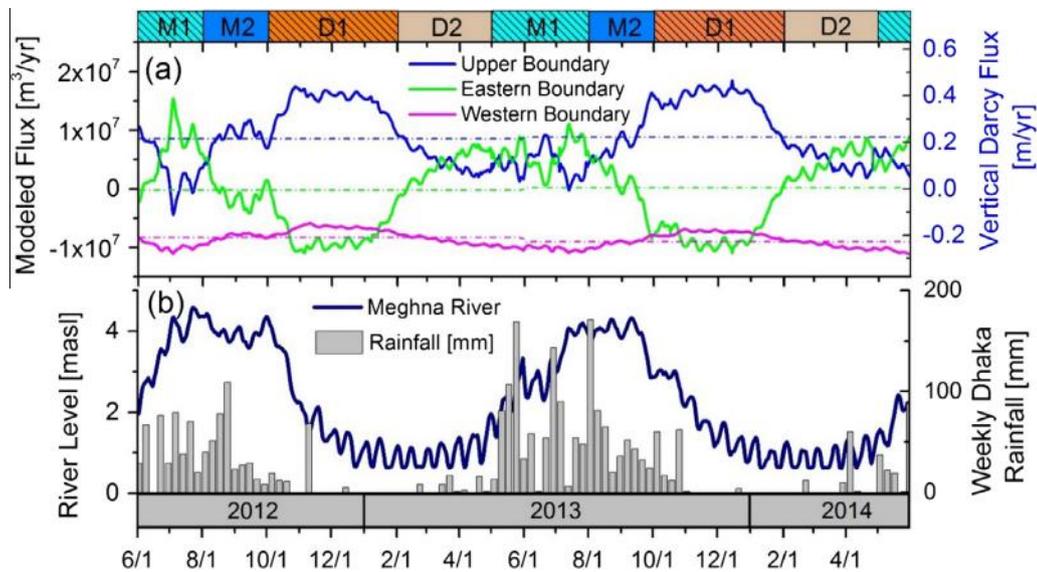


Fig. 11. (a) Modeled recharge (+) and discharge (–) in $6.3 \times 6.3 \text{ km}^2$ deep aquifer study area. Movement across the upper model boundary is expressed as both volumetric flux (left axis) and Darcy flux (right axis). Dashed lines correspond to the 1 year average flux across the boundary. (b) Level of the Meghna River and Weekly total rainfall measured in Dhaka (BMD, 2015).

the drawdown cone of zero (Fig. 11a). The river, however, clearly influences groundwater levels at both shallow and deep depths within several km of its shores. The level of the Meghna River rises ahead of the water table in local aquifers. The river's catchment includes the Meghalaya hills and Sylhet region where annual rainfall is much higher than in Dhaka and substantial rainfall occurs earlier each year (Jakobsen et al., 2005; BMD, 2015). As rainfall continues in the late monsoon, soil becomes saturated and widespread flooding occurs (Fig. 11b). This inland flooding recharges the drawdown cone area and decreases the lateral hydraulic gradient within the study area (Fig. 9b). By the late dry season the lateral gradient once again increases as recharge sources from above have been exhausted.

5.4. Estimating recharge entering the deep aquifer

The smoothly varying, high frequency water level measurements in the eastern Araihaazar study area constrain the shape of the potentiometric surface allowing daily estimates of vertical and lateral recharge to the deep aquifer. The shape of the potentiometric surface varies from linear to strongly quadratic throughout the year (Video S1). Since convergent flow from Dhaka pumping is not the reason for an exponential shape of the potentiometric surface in eastern Araihaazar (Section 3.2), the quadratic shape must be caused by vertical recharge (Eq. (5)). And by extension, a linear hydraulic gradient implies no additional recharge is occurring. We find that vertical recharge to the deep aquifer system in eastern Araihaazar is strongest from the late monsoon through the early dry season and weakest from the late dry season through the early monsoon (Fig. 11a). This is consistent with the timing of the observed peak vertical gradients between the intermediate and deep aquifer on the western end of the modeled domain (Fig. 9c).

In contrast, the modeled lateral recharge from the eastern boundary peaks in the early monsoon (Fig. 11a). The timing of the modeled peak lateral recharge agrees with that of the peak vertical hydraulic gradient measured between the shallow and deep wells on the eastern boundary of the modeled domain (Fig. 9c). The water levels in deep wells lag this early rise. Water levels in shallow wells west of the modeled domain also lag (Fig. 9a). This is why little seasonality was observed in vertical gradients on the western edge of the modeled domain.

Our interpretation is that once widespread flooding occurs in the late monsoon (Fig. 11b), vertical recharge dominates lateral recharge to the aquifer or $w \gg q_0'$ (Figs. 11a and 12d). During the early dry season w continues to increase while the deep aquifer system discharges some of this excess water east towards the river (Figs. 11a and 12a). During the late dry season w decreases to zero as supply from the previous monsoon have been exhausted (Fig. 12b). The steady increase in eastward hydraulic gradient throughout the late dry season is driven by depressurization of the deep aquifer system by urban pumping in Dhaka (Fig. 12b).

5.5. Implications for vertical recharge from shallow aquifers

Dhaka pumping may in some areas already have increased downward recharge of water from shallower, As-rich aquifers into deeper, low-As aquifers. The vertical movement of groundwater is controlled by average vertical hydraulic conductivity (K_v) across the aquifer system. This can be calculated with Darcy's law using modeled vertical Darcy flux, and the observed vertical hydraulic gradients (dh/dz) between the shallow and deep aquifer on the western end of the study area. This was measured from the middle of CW-28 well screen to that of well 24030 (36–205 m depth). Mean annual vertical gradients were 2.61×10^{-2} and 2.75×10^{-2} for 2012–2013 and 2013–2014, respectively (Fig. 9c). Using the Darcy fluxes, K_v equals $2.2(\pm 0.4) \times 10^{-2}$ m/day for both years. The range of possible Darcy fluxes stemming from uncertainty in T within the deep aquifer system are indicated in parentheses. This value of K_v is substantially higher than reported country-wide average estimates of K_v . Ravenscroft et al. (2005) reported a K_v range from 3×10^{-3} to 8×10^{-3} m/day whereas Michael and Voss (2009b) estimated 4×10^{-3} m/day using a combination of basin-wide modeling, C^{14} dating and harmonic averaging of logged lithology with assumed K values.

Vertical hydraulic conductivity can be used to calculate hydraulic anisotropy (K_h/K_v) across the study area. Measured horizontal hydraulic conductivities (K_h) in sands in Araihaazar and surrounding regions typically range from 10 to 40 m/day (Ahmed, 1994; Ravenscroft et al., 2005; Radloff, 2010; Nakaya et al., 2011; Knappett et al., 2012). Combining the upper and lower bounds for both K_v ($2.2 \pm 0.4 \times 10^{-2}$) and K_h , we calculate a range of hydraulic anisotropy from 335 to 2579. This much lower than

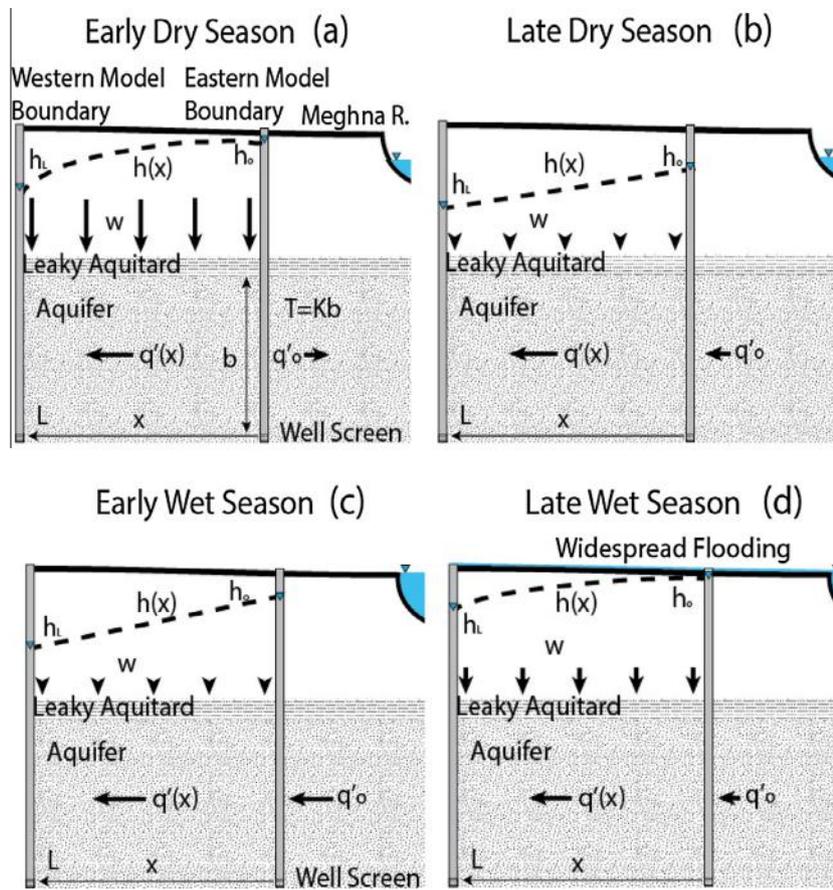


Fig. 12. Conceptual model of the seasonality in modeled recharge and discharge to and from the intensively studied deep aquifer system in eastern Araihaazar.

hydraulic anisotropy of 10,000 estimated by Michael and Voss (2009b), suggesting that the shallow aquifer system in eastern Araihaazar may be more vertically conductive than is typical across the country.

Beyond the effects of hydraulic anisotropy, it is worth pointing out that any downward transport of shallow high As groundwater is likely to be retarded as it passes through brown, oxidized sand. A push–pull test conducted within brown, oxidized sands at 65 m depth in western Araihaazar measured an average As retardation of 14 (Radloff et al., 2011). A recent study in Vietnam documented the conversion of orange Pleistocene sand to gray, reducing sand and a concomitant release of As. This study estimated retardation of the high As pore water front to be 16–20 times slower than the average linear groundwater velocity carrying water from reducing aquifers (van Geen et al., 2013). In addition to direct transport of As, DOC from shallow aquifers may convert brown, oxidized sand to reduced gray sands and liberate *in situ* As through reductive dissolution of metal oxides (Harvey et al., 2002; McArthur et al., 2008; Neumann et al., 2010; Mailloux et al., 2013; van Geen et al., 2013). Using a range of retardation factors from 14 to 20 under, current (local) conditions, we estimate the average downward movement of the high-As zone in eastern Araihaazar conditions to be 2–5 cm/yr, although it is likely to be higher in western Araihaazar, closer to the pumping center. This moderate rate of contamination helps explain why no wide-scale contamination of low-As aquifers >90 m deep over time has been documented to date in Araihaazar or much of Bangladesh (van Geen et al., 2007, 2016; Ravenscroft et al., 2013, 2014).

This rate of downward flow will likely increase with a continued increase in Dhaka municipal pumping rate. There are long

term records of the rates of municipal pumping going back to the 1960s (DWASA, 2012). The increase in DWASA pumping rates since 1990 can be fit with either an exponential or a linear curve with R^2 values of 0.99 and 0.98, respectively. The exponential and linear curves predict a doubling time of 12 and 29 years, respectively. The regional drawdown cone model (Hantush and Jacob, 1955) predicts that downward advective velocity will double with a doubling of the Dhaka pumping rate. Therefore this rate of downward movement estimated in the present study for eastern Araihaazar is likely to increase as the rate of Dhaka pumping increases. The system of aquifers is, however, more heterogeneous than assumed by the regional model so there is not a predictable outcome of increased pumping by Dhaka. For example the shallow aquifer may become perched such that vertical recharge rates will no longer depend on decreasing pressure in the deep aquifer system. Further, if the deep aquifer system was dewatered, this would temporarily slow the expansion of the drawdown cone as water came from gravity drainage from pore spaces.

5.6. Implications for recharge from rivers

Urban pumping may capture recharge from rivers and potentially directly mobilize As bound in sediments lining rivers like the Meghna River (Datta et al., 2009; Jung et al., 2012). Past studies of the Dhaka drawdown cone suggested the lateral extent of the (then shallower) drawdown cone would be limited by geology and local rivers bordering Dhaka (Ahmed et al., 1998). These studies noted that the drawdown cone appeared to be confined by the Shitalakshya and Buriganga Rivers (Fig. 1). The present work and another recent study (Hoque et al., 2007) demonstrate that this

is not the case as the Dhaka drawdown cone now extends far to the east of the Shitalakshya River. Further, Shamsudduha et al. (2009) demonstrated the drawdown also extends south of the Buriganga River (Fig. 1). The depressurized regional aquifers therefore raises the prospect of accelerated recharge through river banks. Given the high levels of As accumulating in river bank sediments (Datta et al., 2009; Jung et al., 2012) and the reactivity of organic carbon in freshly deposited river sediments (Postma et al., 2010), a net flow reversal caused by Dhaka pumping could be accompanied by an increase in As concentrations in adjacent aquifers

6. Conclusions

If Dhaka pumping continues to increase many villagers will lose access to low-As drinking water and, over the next decade, currently low-As parts of the aquifer system may gradually become contaminated. Although this has not been documented yet, downward movement of high-As water may eventually increase As concentrations in deeper wells that are currently low in As. Arahazar's 11,639 private wells placed at shallow depths (30–90 m) and 139 community wells placed at intermediate depths (90–150 m) would likely be affected first. As of 2014, 2209 (19%) of private wells (30–90 m) and only 3 (1.4%) of intermediate depth community wells exceeded the Bangladesh drinking water limit for As of 50 ppb. The benefits and losses of continued Dhaka pumping are unequally distributed: the urban population benefits from the water supply whereas rural areas not served by municipal water are those whose resource is affected in both the short (declining water levels) and possibly in the long (migrating As) term. DWASA has announced plans to gradually switch to treated surface water as a source of drinking water. This would lead to a recovery of hydraulic heads in the deep aquifer. Therefore, contamination of the surrounding intermediate and deep aquifers with As may therefore still be averted.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.jhydrol.2016.05.035>.

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