

1 **Abstract**

2 The high-degree of spatial variability of dissolved As levels in shallow aquifers of the Bengal
3 Basin has been well documented but the underlying mechanisms remain poorly understood. We
4 compare here As concentrations measured in groundwater pumped from 4700 wells <22 m (75 ft)
5 deep across a 25 km² area of Bangladesh with variations in the nature of surface soils inferred
6 from 18,500 measurements of frequency-domain electromagnetic induction. A set of 14 hand
7 auger cores recovered from the same area indicate that a combination of grain size and the
8 conductivity of soil water dominate the electromagnetic signal. The relationship between pairs
9 of individual EM conductivity and dissolved As measurements within a distance of 50 m is
10 significant but highly scattered ($r^2=0.12$; $n=614$). Concentrations of As tend to be lower in
11 shallow aquifers underlying sandy soils and higher below finer-grained and high conductivity
12 soils. Variations in EM conductivity account for nearly half the variance of the rate of increase
13 of As concentration with depth, however, when the data are averaged over a distance of 50 m
14 ($r^2=0.50$; $n=145$). The association is interpreted as an indication that groundwater recharge
15 through permeable sandy soils prevents As concentrations from rising in shallow reducing
16 groundwater.

1 **1. Introduction**

2 It took a small group of dedicated scientists a decade to convince the world that elevated As
3 concentrations in groundwater pumped from millions of wells across the Bengal Basin poses a
4 serious health threat (Chakraborty and Saha, 1987; Dhar et al., 1997). After another decade of
5 intense field and laboratory research conducted by many more researchers, the processes that led
6 to elevated As concentration in groundwater of the region remain poorly understood. One reason
7 for limited progress is the extreme degree of spatial variability of the geology of a large fluvio-
8 deltaic system such as the Bengal Basin (BGS/DPHE, 2001; Yu et al., 2003; Ravenscroft et al.,
9 2005). This variability has made it difficult to generalize detailed observations of groundwater
10 and aquifer solids obtained from a limited number of sites to a broader region (e.g. BGS/DPHE,
11 2001; Harvey et al., 2002; McArthur et al., 2004). Despite the underlying geological
12 complexities, and therefore hydrological complexities, the interaction of potentially multiple
13 factors controlling As concentrations in groundwater must be understood as it is likely to
14 determine the sustainability of aquifers in the region that are presently low in As and offer the
15 best hope of reducing the exposure of the population to As in the short term (van Geen et al.,
16 2002; 2003a; Ahmed et al., 2006).

17
18 In an attempt to decompose the groundwater As problem into more tractable questions, the
19 present study focuses on the processes that regulate As concentrations in shallow aquifers of
20 relatively recent (<10 kyr, i.e. Holocene) age. The field observations extend over a 25 km² area
21 of Araihasar upazila where the distribution of As in the groundwater has been mapped at
22 unprecedented resolution by sampling several thousand tubewells (Fig. 1a; van Geen et al.,
23 2003b). When considering only shallow wells, defined here as wells <22 m (75 ft) deep (metric
24 and imperial units of depth are listed because the latter are more widely used in Bangladesh),
25 Araihasar marks a transition zone between the coastal region to the south, where very few wells
26 meet the 50 µg/L Bangladesh standard for As in the drinking water, and inland areas to the north
27 where most shallow wells meet the 10 µg/L WHO guideline for drinking water As (BGS/DPHE,
28 2001). Within Araihasar and other similarly affected regions of Bangladesh, average
29 groundwater As concentrations in shallow aquifers generally increase with depth at any
30 particular location (BGS/DPHE, 2001; Harvey et al., 2002; Horneman et al., 2004). The
31 relationship is highly scattered when extended over larger areas, however. Even within the
32 confines of our 25 km² study area, there are numerous tubewells that are elevated in As and
33 others that are low in As throughout the 8-22 m depth range (Fig. 2). The present study is an
34 attempt to shed some new light on the origin of this spatial variability by applying, to our
35 knowledge for the first time in Bangladesh, an established and simple geophysical surveying
36 technique, frequency-domain electromagnetic induction, to relate spatial variations in the nature
37 of surface soil properties to As concentrations in shallow groundwater.

38
39 Frequency-domain electromagnetic (EM) induction has been used in hydrology to map
40 percolation of water in the vadose zone, to estimate the extent and internal structure of shallow
41 aquifers, and to determine the extent of groundwater contamination (McNeill, 1980a and 1990;
42 Cook et al., 1989; Pellerin, 2002; Pettersson and Nobes, 2003). EM conductivity has also helped
43 classify soil types in a mid-Atlantic coastal plain (Anderson-Cook et al., 2002). A number of
44 studies have shown that hand-held EM instruments deployed by walking through cultivated
45 fields can also be used to determine the spatial variability of soil salinity (McNeill, 1986;
46 Hendrickx et al., 1992; Lesch et al., 1995; Vaughan et al. 1995; Doolittle et al., 2001; Triantafilis

1 et al. 2002). It has, however, typically been difficult to disentangle the direct contribution of
2 fine-grained particles to EM conductivity from an indirect contribution due to the accumulation
3 of salt by evaporation at the surface of relatively impermeable deposits (Williams and Hoey,
4 1987; Cook et al., 1989; 1992; Doolittle et al., 1994; Humphreys et al., 1995, Johnson et al.,
5 2001, Triantafilis et al., 2002). In addition to salt content and grain-size, some studies have
6 shown that variations in soil moisture can influence the EM conductivity signal (e.g Waive et. al.,
7 2000) whereas, in a different setting, Banton (1997) observed little difference in EM response
8 between wet and dry conditions.

9
10 In this paper, we compare the spatial distribution of over 18,000 EM induction measurements in
11 open fields of Araihasar with As concentrations in groundwater sampled from 4700 shallow
12 wells (<22 m or <75ft deep) from the same area. The next section of this paper (Section 2)
13 describes the geological setting of the study and the spatial distribution of arsenic in shallow
14 groundwater. Section 3 reviews the principles of EM induction and describes how the surveys
15 were conducted. Collection and analysis of 14 soil profiles obtained with a hand auger to
16 calibrate the EM response are also presented. In Section 4, the general pattern of EM variations
17 in the study area is described and compared to soil profiles. Section 5 leads to a simple linear
18 regression model of shallow groundwater As as a function of both depth and EM conductivity.
19 Section 6 offers an explanation for the observed patterns that draws from a recent demonstration
20 by Stute et al. (2007) of the influence of local variations in recharge on the As content of shallow
21 Bangladesh aquifers.

22 23 **2. Hydrogeological Setting**

24 Erosion of mountains surrounding the Bengal Basin has deposited thick layers of clay, silt, sand,
25 and gravel across much of Bangladesh over the past 7 kyr of the Holocene (Morgan and McIntire,
26 1959; Umitsu, 1993; Goodbred and Kuehl, 2000; BGS and DPHE, 2001; Goodbred et. al., 2003).
27 Our study area within Araihasar straddles a transitional region that extends from the uplifted
28 Madhupur Tract to the northwest, where only surface soils contain recently deposited material, to
29 the active floodplain of the Meghna River to the southeast where up to 150 m of Holocene
30 sediment accumulated during the Holocene (van Geen et al., 2003b; Zheng et al., 2005). The
31 Old Brahmaputra River running through the study area today is a modest stream that stops
32 flowing during the dry season (Fig. 1a & 1b), but there are indications of a more powerful river
33 passing through the area that may have shifted its course in the 18th century along with the main
34 channel of the Brahmaputra River (Fergusson, 1863; Brammer, 1996; Weinman et al., in press).

35
36 Time series of river stages and groundwater levels as well as measurements of vertical hydraulic
37 gradients at six multilevel well sites in Araihasar over two years show a seasonal pattern in
38 groundwater levels that mimics the river stages, with different time lags during the rising and the
39 falling periods (Zheng et al., 2005; Stute et al., 2007). At the end of the dry season, there is a
40 slight delay in the response of groundwater to rapidly rising water levels in rivers that is
41 accompanied by an upward hydraulic head gradient, suggesting a lateral input of water from
42 rivers to aquifers. There are also short periods at the beginning of the wet season when
43 groundwater levels rise above the river and vertical hydraulic head differences point downward,
44 probably due to recharge with precipitation. Whereas during the wet season (June – October),
45 groundwater and river levels are comparable, the drop in groundwater levels considerably lags
46 the decline in river levels into the dry season. Such seasonal differences indicate that recharge

1 occurs rapidly over a larger area through bank filtration, flooding, and infiltration of rainwater,
2 while discharge occurs at fewer locations such as local surface water bodies and by localized
3 evapotranspiration. Groundwater flow during the dry season is also likely to be affected by
4 irrigation pumping during the 2-4 month period when mechanized tubewells are used throughout
5 the area to flood rice paddies with groundwater. Similar observations have been reported
6 elsewhere in Bangladesh (BGS/DPHE, 2001), including a study area in Munshiganj, ~30 km
7 southwest of Araihasar (Harvey et al 2002; 2006; Klump et al., 2006). In Munshiganj, shallow
8 aquifers are semi-confined because of extensive surficial deposits of clay and fine clay. Whereas
9 fine-grained surface deposits are observed in Araihasar, there are also large areas where sandy
10 deposits extend to the surface and are not capped by an impermeable layer (Horneman et al.,
11 2004; Stute et al., 2007; Weinman et al., in press). Direct rainfall, flooding, and seepage from
12 irrigated fields and surface water bodies like ponds may therefore constitute a larger component
13 of recharge in Araihasar compared to Munshiganj (Harvey et al., 2006; Stute et al., 2007).

14 15 **3. Methods**

16 **3.1 EM Conductivity Survey**

17 The geophysical survey of the study area was conducted with a Geonics[®] EM31 instrument
18 (McNeill, 1980a, 1990). The EM31 consists of a transmitter coil radiating an electromagnetic
19 field at 9.8 kHz and a receiver coil located at opposite ends of a 3.7-m long boom. By inducing
20 an eddy current in the ground, the primary field generates a secondary electromagnetic field that
21 is recorded by the receiving coil. The intensity of the secondary field increases with the
22 conductivity of the ground. This conductivity is a function of the concentration of ions dissolved
23 in soil water as well as exchangeable ions at the solid/liquid interface. The secondary field
24 generated below the soil surface diminishes with depth. The penetration of the signal depends
25 on the separation between the transmitting and receiving coils, the transmission frequency, and
26 the coil orientation (McNeil, 1980a). When the boom is held at waist height horizontally, which
27 corresponds to a vertical dipole orientation, 50% of the signal is generated in the upper 90 cm of
28 the soil, 23% between 90 and 180cm, and the remaining 27% by the conductivity of layers below
29 180 cm depth (McNeil, 1980a; Doolittle, 2001).

30
31 Nearly 18,500 EM31 readings were collected at waist level in Araihasar, primarily during the
32 dry months of January 2001, January 2002, and March-June 2002 (Figs. 1a & 1b). Flooding
33 precluded data collection during the wet season. At the beginning of each day, the instrument
34 was calibrated following the standard procedure described in the operating manual. The distance
35 between sequential measurements along a particular transect ranged from 5 to 10 wide steps, i.e.
36 ~4 to 8 m. The reproducibility of readings at a single location was generally within 0.5 mS/m.
37 Each determination rounded to the nearest unit of mS/m was entered by hand in the field on a
38 Compaq Pocket PC connected to a Global Positioning System (GPS) receiver using ESRI[®]
39 ArcPad software. EM conductivity measured along the edge of flooded rice fields was typically
40 ~1 mS/m higher than over adjacent dry areas. Coverage is limited to open areas to avoid
41 interference from corrugated iron plates used in the villages for building and fences. EM data
42 were also not collected within 10-20 m of overhead power lines.

3.2 Collection and Analysis of Soil Samples

At 14 locations spanning the spectrum of EM conductivities measured in Araihaazar (Figs. 1a & 1b), a total of 112 soil samples were collected with a hand auger to a depth of 200 cm. EM conductivities were measured at the auger sites at the time of soil sample collection. The particle size distribution and the electrical conductivity of sediment slurry were determined every 25 cm. Samples were disaggregated in distilled water and washed through a 63 μm sieve to determine the sand fraction. A Micromeritics SediGraph 5100 was used to quantify the silt and clay fraction down to 0.8 μm . For slurry conductivity measurements, wet soil (~2 g) was diluted in 15 mL of deionized high-purity water delivered by Milli Q system and allowed to equilibrate overnight. An Orion 105Aplus conductivity meter with a 011050MD conductivity probe, calibrated with a 1413 μS standard solution (Orion 011007), was then used to measure the conductivity of the slurries. The water content of soil samples, tightly packed in plastic containers at the time of collection, was measured by drying in an oven at 65°C for 24 hours and ranged from 0.5 % by weight in the case of sands to 30% in clays.

3.3 Hydraulic head measurement

Stute et al. (2007) reported monthly variations in hydraulic head for nests of wells distributed across the study area as well as a confluent of the Old Brahmaputra River (Fig. 1b). These measurements were extended by second year to cover 2004-2006. Hydraulic heads in the piezometric wells and river level were measured by a water level meter (Solinst 101) on a bi-weekly to monthly basis. Absolute elevations of the top of one well from each multilevel well site and a reference mark at the river monitoring station were determined in January 2003 using a Differential Global Positioning System (DGPS) static survey technique (Zheng et al., 2005). Elevation of the top of other wells relative to the reference well at each multilevel well site, was measured visually within ± 1 mm by leveling with a transparent flexible tube filled with water (Zheng et al., 2005).

4. Results

4.1 Distribution of EM conductivity

EM conductivities measured in open areas of Araihaazar span a wide range of 3 to 75 mS/m and average 18 ± 8 mS/m ($n=18,530$). The histogram of EM conductivities is slightly skewed towards higher values, with a median of 17 mS/m and a variance of 48 mS/m (Fig. 3). Nearly 99% of the measurements are in the 10-40 mS/m range, however. The largest region where elevated conductivities (30-50 mS/m) were consistently recorded is located in the northwestern part of the study area (Fig. 1b). There are also two smaller patches of elevated conductivities ~1.5 km to the northeast and south of this region, respectively. A large 0.5 to 1 km-wide swath of moderately high EM conductivities (20-30 mS/m) runs north-south through the eastern portion of the study area. This is also a region where the As content of the vast majority of wells <22 m (75 ft) deep exceeds 100 $\mu\text{g/L}$ (Fig. 1a).

At the other end of the spectrum of EM measurements, a well defined swath of low conductivity (<13 mS/m) runs diagonally along the northwestern boundary of the study area (Fig. 1b). Several villages located within this region are populated with shallow wells that are consistently low in As, but there are also patches within this area containing wells that are predominantly elevated in As (Fig. 1a). The southern diagonal alignment of villages with wells predominantly low in As also appears to be associated with generally low conductivities in surrounding fields.

1 **4.2 Profiles of Soil Properties**

2 **4.2.1 Conductivity of Soil Slurries**

3 EM conductivities range from 3 to 35 mS/m at the 14 sites where auger cores were collected (Fig.
4 1b). The conductivity of slurries of the underlying soil measured with a conductivity probe,
5 referred to hereon as slurry conductivity, range from 5 to 60 mS/m after dilution. For ease of
6 viewing, the profiles of slurry conductivity are subdivided into three groups. Slurry
7 conductivities average 12 ± 12 mS/m ($n=112$) and are essentially uniform with depth for the 6
8 profiles in the lowest category of EM conductivities (3-10 mS/m; Fig. 4a). This is also the case
9 for 1 out of 4 profiles from sites in the intermediate EM conductivity range (11-15 mS/m; Fig.
10 4a). The other 3 profiles in this category, however, show 2 to 6-fold increases in slurry
11 conductivity in the deepest sample at 200 cm. A similar increase in conductivity at depth is
12 observed for 3 out of 4 profiles in the highest category of EM conductivities (20-35 mS/m; Fig.
13 4a). In this category, slurry conductivities in the upper 150 cm of the soil are also higher (14 ± 8
14 mS/m, $n=14$) than for the two groups of sites showing lower EM conductivities.

16 **4.2.2 Grain-size**

17 The nature of the soils sampled with the hand auger ranged from thick layers of sticky clay to
18 deposits of coarse sand similar to aquifer material from which groundwater is extracted at greater
19 depths. Although the contribution of intermediate-sized silts also varied significantly (2-72 %),
20 the grain-size data can be conveniently summarized on the basis of the clay fraction ($< 2 \mu\text{m}$)
21 profiles. One justification is that the sand and silt fraction of soils are poor conductors (McNeill,
22 1980b). Soil conductivity is therefore largely determined by the proportion of clay. With the
23 exception of a few horizons, the clay content of soils in the groups of profiles with low and
24 intermediate EM conductivities ranges from 1 to 20% (Fig. 4b). The clay content is $< 5\%$ in 21
25 out of 48 samples in the low EM category but in only 6 out of 32 samples in the intermediate
26 category. In contrast, the clay content of all samples collected from sites in the highest EM
27 category is always $> 10\%$ and frequently exceeds 50% (Fig. 4b). At the site where the highest
28 EM conductivity of 35 mS/m was measured, the clay content of the soil in 7 out of 8 samples is
29 $> 60\%$. The core collected from the site with the lowest EM value within the high EM category
30 (20 mS/m) is also the only one that contained some silty sand. The correlation between slurry
31 conductivity and clay fraction for individual auger samples is poor, however ($r^2=0.34$, $n=112$).
32 This is because slurry conductivities generally increase with depth whereas the clay fraction is
33 often higher towards the surface (Fig. 4).

35 **4.3 Water levels in Araihasar**

36 Hydraulic heads are displayed as box plots of the difference in head between the shallowest (< 15
37 m) well from five multi-level well sites and the river for the pre-monsoon (February-May),
38 monsoon (June-September), and post-monsoon (October-January) period (Fig. 5a). Positive
39 head differences from each monitoring site indicate potential groundwater flow towards the river
40 and vice versa. The average water level in the shallowest well from each site remains close to
41 that of the river during the monsoon at three of the sites (sites A, C, and F). The difference in
42 water levels is more variable at the two other sites (sites C and E) and on average below that of
43 the confluence of the Old Brahmaputra River, suggesting more local control. Groundwater levels
44 in shallow wells at four of the sites remain at or above that of the river during the four months
45 preceding the monsoon (Fig. 5a). The one exception is Site F where irrigation is particularly
46 intense (Fig. 1a) and average water levels in shallow aquifers remain below that of the river (Fig.

1 5a). At all five sites, however, average hydraulic gradients over the entire two year period point
2 towards the river (Fig. 5b).

3
4 Although there are many private wells in our study area (Fig 1a), we are not confident in drawing
5 a regional hydraulic head contour map because we were unable to obtain detailed hydraulic and
6 elevation data from these wells. Relying on these private wells would also have introduced a
7 bias because they cover only 40% of the total area and are located within villages, which are
8 typically elevated relative to surrounding fields. Also, private wells are equipped with suction
9 handpumps, which are hard to remove and re-install because of corrosion of bolts. In addition,
10 measuring hydraulic heads with an electric tape can cause microbial contamination of a well.
11 Finally, resistance from the well owners and the amount of time needed (hours) to open and re-
12 seal each well prevented us from collecting these data simultaneously over a large area. Our
13 nests of monitoring wells are more easily accessible and some are located in the fields, but the
14 distances of these nests from each other are much larger than the likely scale of spatial variability
15 of hydraulic head.

16 17 **5. Discussion**

18 **5.1 EM Conductivity and Sediments Properties**

19 From first principles, McNeil (1980a) showed that the contribution from different depths to EM
20 conductivity can be calculated as $S_A = S_1 [1 - R_V(Z_1)] + S_2[R_V(Z_1) - R_V(Z_2)] + \dots + S_N R_V(Z_N)$,

21 where $S_1 \dots S_N$ are soil conductivities at depths $Z_1 \dots Z_N$, and $R_V(Z) = \frac{4Z}{(4Z^2 + 1)^{3/2}}$ is the

22 cumulative response function specifying the contribution from each depth, expressed as a
23 fraction of the distance separating the emitting and receiving coils. We make here no attempt to
24 calibrate the EM31 signal in any absolute sense, nor do we try to separate the contribution to EM
25 conductivity of ions present in soil water and exchangeable ions at the solid/solution interface
26 (Rhoades JD, 1981; Rhoades and Corwin, 1981; Rhoades et al, 1989; Cook et al., 1989). Instead,
27 we compute a depth-weighted slurry conductivity and a depth-weighted clay fraction from
28 different soil layers by applying the formulation of McNeil (1980a) separately to the measured
29 slurry conductivities and to the clay fractions at every auger site. Contributions from different
30 depths were corrected according to the height (0.5m) of the instrument above the ground. EM
31 conductivities measured over the 14 auger sites broadly increases with the integrated slurry
32 conductivity (Fig. 6a). EM conductivities also increase fairly systematically as a function of the
33 depth-weighted proportion of clay in the soil (Fig. 6b). Linear regression shows that depth-
34 weighted slurry conductivity ($r^2=0.85$, $n=14$) and depth-weighted clay fraction ($r^2=0.85$, $n=14$)
35 correlate with EM conductivities measured at the surface, and this despite the significant
36 contribution to the EM signal of layers below 2 m depth.

37
38 It is difficult to distinguish the contributions of the clay fraction and the conductivity of soil
39 water to the EM signal. However, previous studies have shown that the controlling factor in
40 some areas is clay content (Dalgaard et al., 2001; Durlleser, 1999; Hedley et al., 2004;
41 Triantafilis and Lesch, 2005). In a setting similar to Bangladesh, Kitchen et. al. (1996) observed
42 strongly reduced EM conductivity over sand deposits dating from the 1993 flood of the Missouri
43 River compared to surrounding finer soils. Doolittle et al. (2002) also located sub-surface sand
44 blows in southeastern Missouri by EM conductivity. In Australia and in the US, EM
45 conductivity surveys have been used to optimize rice production by identifying areas where

1 irrigation water is lost to infiltration through sandy deposits (Beecher et.al., 2002; Anderson-
2 Cook et.al., 2002). What matters in the present context is that the auger cores show that the EM
3 conductivity survey of Araihasar produced an extensive map of a property that is closely related
4 to the clay content, and therefore the permeability, of surface soils.

5.2 Geostatistics of EM Conductivity

7 The experimental variogram provides a convenient graphical representation of the continuity, or
8 roughness, of spatial data (Cressie, 1993; Robertson, 2000; Webster and Oliver, 2001). It is

9 calculated using the equation $\gamma(h) = \frac{1}{2N(h)} \sum (Z_i - Z_{i+h})^2$ where $\gamma(h)$ is semivariance for

10 interval distance class h , Z_i is measured sample value at a point h , Z_{i+h} is measured sample value
11 at a point $i+h$ and $N(h)$ is the total number of sample couples for the lag interval h . To obtain the
12 variogram of EM conductivity in Araihasar, distances up to 1000 m separating two
13 measurements were considered and binned in 10-m intervals. The results show that the variance
14 of pairs of EM measurements increases steadily from 0.1 (mS/m)² for sites 10 m apart to ~55
15 (mS/m)² at a distance of 400 m (Fig. 7). This quantitatively confirms our experience from the
16 field that a spatial resolution of ~10 m is generally sufficient to capture variations in EM
17 conductivity >1 mS/m. The increase in variance between pairs of determinations up to a distance
18 of 400 m is consistent with the ~500 m scale of several areas of consistently high and low
19 conductivity distributed across the study area (Fig. 1b). An exponential model was fitted to the
20 variogram by minimizing the sum of squared residuals between the model and the observations
21 (RSS = 2266; $r^2 = 0.83$; Fig. 7). The model was used to contour the EM conductivity data and
22 visually enhance the main features of the survey by ordinary block kriging. Ordinary kriging
23 estimates optimally-weighted averages for unsampled locations of an irregularly spaced data set
24 using knowledge about the underlying spatial relationships (Cressie, 1993, Robertson, 2000). A
25 cross-validation comparing estimated and actual EM conductivities, [$y = (1.01 \pm 0.01) * x -$
26 (0.17 ± 0.05)], where y = predicted EM conductivity and x = actual EM conductivity] shows that
27 kriging adequately captures the main features of the variability in the nature of surface soils, and
28 therefore permeability, with a standard error for predicted EM of ~2 mS/m based on the standard
29 deviation of residuals.

5.3 Relation between EM conductivity and As in groundwater

32 The contour map of EM conductivity facilitates visual comparison with the distribution of As in
33 shallow wells because the two data sets generally do not overlap (Fig. 8). Merging of the two
34 data sets shows, for instance, that interruptions of the diagonal swath of mostly low-As wells
35 across the northwestern portion of the study area by clusters of high-As wells occur precisely in
36 areas of elevated EM conductivity. Similarly, two clusters of generally low As concentrations to
37 the south are closely delimited by areas of elevated EM conductivity. Overall, the average As
38 content of shallow wells located within the 10 mS/m contour is 43 ± 67 $\mu\text{g/L}$ ($n=606$) whereas the
39 average As content of wells located outside the same contour is 120 ± 123 $\mu\text{g/L}$ ($n=2199$). A
40 different way to express the contrast between the two areas is that 73% of shallow wells located
41 within the 10 mS/m contour contain <50 $\mu\text{g/L}$ As, whereas this is the case for only 36% of wells
42 outside the same contour. These observations suggest that shallow tubewells that meet the
43 Bangladesh standard for groundwater of 50 $\mu\text{g/L}$ As are unlikely to be found in areas where
44 surface soils contain >10% clay (Fig. 4).

1 For a more quantitative comparison between groundwater As and EM conductivity data, we
2 return to the actual EM data instead of extrapolated values by comparing the As content of each
3 shallow well to the closest EM conductivity measurement. To determine the relevant spatial
4 scales to consider, as determined by the ratio of horizontal to vertical flow, we estimate the
5 distance over which recharged groundwater has to travel to reach a depth of 15 m (over two-
6 thirds of shallow household wells considered in this study are up to 15 m deep). For lateral flow,
7 we start from annually-averaged relative hydraulic head differences between the multilevel sites
8 and the Old Brahmaputra River (Fig. 5b). In the case of sites A and C, we use the actual distance
9 of discharge to the river to calculate lateral gradients in average hydraulic head (100 and 300,
10 respectively; Table 1). For sites of E and F, we estimate the shortest distance to a local discharge
11 area to be on the order of 150 m rather than actual distances to the Old Brahmaputra River (Table
12 1). Vertical hydraulic gradients between the two shallowest wells at all four sites were
13 previously reported by Stute et al. (2007). Using the ratio between horizontal and vertical
14 gradients and assuming a vertical anisotropy (K_x/K_z) of 10 (Freeze and Cherry, 1979), distances
15 over which recharged groundwater could have traveled to reach a depth of 15 m were then
16 calculated. The results shows that groundwater from 15 m depth may have been advected
17 laterally by 50 m at site F and by as much as 700 m at site E (Fig 9, Table 1). This is just an
18 order of magnitude estimate because the hydraulic gradient between wells nests and the river is
19 not necessarily linear.

20
21 On the basis of this simple calculation, we start by comparing the As content of each shallow
22 well to the closest EM conductivity measurement obtained within a distance of 50 m. Linear
23 regression shows a weak but statistically significant relationship ($R^2 = 0.12$, $p < 0.001$) for 614
24 paired measurements corresponding to an increase in As concentrations as a function of EM
25 conductivity (Fig. 10a). The relationship remains significant but weakens further when paired
26 measurements separated by larger distances are considered (Table 2a). Averaging EM data
27 around the subset of 504 wells surrounded by at least five measurements yields a relationship
28 that accounts for a larger proportion of the variance ($R^2 = 0.22$). The magnitude of standard
29 deviations for five or more EM conductivity measurements within 50 m of a well shows that part
30 of the scatter is due to the spatial variability of EM conductivity (Fig. 10b). The outcome of
31 these regressions does not change markedly when considering a smaller or larger minimum
32 number of EM measurements within a 50 m radius (Table 2b).

33
34 Vertical profiles of As for nests of monitoring wells across the study area suggest that EM
35 conductivities could instead be compared to the depth gradient of As concentrations in shallow
36 aquifers rather than absolute As concentrations. Concentrations of As start from <50 $\mu\text{g/L}$ in the
37 shallowest monitoring well at each site and typically increase with depth, gradually at sites
38 associated with low EM conductivities and more rapidly at high conductivity sites (Fig. 11).
39 Least squares regression of As as a function of depth, with a non-negativity constraint imposed
40 on the intercept, shows that the slope of this relationship increases systematically as a function of
41 average EM conductivity within a radius of 50 m of each of these sites (Fig. 12a). The slope of
42 the As-depth relationship increases nearly five-fold across the 7-20 mS/m range in average EM
43 conductivity at these five sites.

44
45 When considering household wells located within 50 m of each of the five nests of the wells, the
46 depth gradient in As concentrations is not as well defined, at least in part because household

1 wells are typically deeper than the shallowest of the monitoring wells (Fig. 11). The scatter of
2 the depth trends in As is also considerably larger for household wells, possibly because of errors
3 in reported well depths as well as lateral spatial heterogeneity. Despite these limitations, the data
4 show that the slope of the As-depth relationship based on household wells also generally
5 increases as a function of EM conductivity (Fig 12a). It is unclear, however, why the depth
6 gradients calculated from the household wells are a factor of three below the gradients calculated
7 from the monitoring wells.

8
9 The results from the area surrounding the five nests of wells justify a similar comparison of As
10 depth gradients and EM conductivity within the larger data set. A total of 145 circles of 50 m
11 radius within the study area contain at least 5 household wells and at least 5 EM conductivity
12 measurements (Fig. 1b). Although there is considerable scatter, the relationship between EM
13 conductivities and the As-depth gradient is highly significant ($p < 0.001$) and accounts for 50% of
14 the variance (Fig. 10c). Whereas the proportion of the variance that can be accounted for
15 depends on the number of wells or EM measurements used for averaging, the relationship
16 between As-depth gradients and EM conductivity itself is not very sensitive (Table 3). In the
17 next two sections we examine why the rate of increase of As concentrations with depth in
18 shallow aquifers of Araihasar might depend on the clay content of surface soil inferred from EM
19 conductivity.

20 21 **5.4 Relation between EM conductivity and groundwater age**

22 Independently of its relation to As, it is worthwhile to compare the distribution of EM
23 conductivity with groundwater ages at the five nests of wells within the study area where the ^3H -
24 ^3He dating technique was applied (Fig. 7). Stute et al. (2007) showed that the time elapsed since
25 recharge increases with depth at all five sites, but at a rate that varies considerably from one nest
26 of wells to the other. At the two sites located within an area of low EM conductivity (sites C and
27 F), the age of groundwater interpolated to a depth of 12 m (40 ft) is 3 to 4 years. In contrast, the
28 age of groundwater at the same depth at the two sites surrounded by regions of high EM
29 conductivity (sites A and B) is 18 to 19 years. Considering recharge rates estimated by Stute et
30 al. (2007) from the vertical $^3\text{H}/^3\text{He}$ age gradient for the shallowest two samples at each well nest
31 confirms that recharge is correlated with EM conductivity (Fig. 10b) and shows that sandy
32 deposits that extend essentially to the surface are recharged an order of magnitude faster than
33 shallow aquifers that are capped by a relatively impermeable layer of fine-grained sediments.

34
35 Similar conclusions were drawn from a combination of recharge estimates and electromagnetic
36 measurements in southeastern Australia, albeit under considerably drier conditions than in
37 Bangladesh (Cook et al., 1992). The same study predicts an exponential relationship between
38 recharge and EM conductivity that may be consistent with our observations, even if a linear
39 relationship could equally be inferred from our limited data (Fig. 10b). The observations from
40 Araihasar are more directly related to recent work conducted in the Himalayan foothills of India
41 (Israel et al., 2006). In that study, the penetration of ^3H injected in surface soil by infiltration of
42 precipitation and return seepage from irrigated fields was shown to increase with the
43 permeability of the soil, as determined by electrical resistivity measurements. Our observations
44 from Araihasar provide further quantitative evidence that variations in surface permeability
45 strongly influence the rate of local recharge of shallow fluvio-deltaic aquifers.

1 To what extent are shallow aquifers of Araihasar affected by irrigation pumping? Mechanized
2 irrigation wells in the area are typically constructed with filters that extend from ~6 to 21 m (20
3 to 70 ft) depth. A total of 125 mechanized irrigation wells were identified during a systematic
4 survey of a 14 km² portion of the study area (Fig. 1a), approximately 40% of which is used for
5 growing rice according to satellite imagery (van Geen et al, 2003). A crop of rice requires ~1 m
6 of irrigation water per year (Meharg and Rahman, 2003; Huq et al., 2006). This requirement,
7 combined with the density of irrigation wells and typical pumping rates of 20-30 L/s (Harvey et
8 al., 2006; van Geen et al. 2006), is consistent with irrigation wells operating 3-4 hours each day
9 during four months of the dry season. Stute et al. (2007) concluded that vertical recharge rates in
10 the same study area range from 0.05 to 1 m/yr using two independent methods: by measuring
11 vertical head gradients and hydraulic conductivities and by ³H-³He dating of shallow
12 groundwater. Comparison with the water requirement for growing rice indicates that the scale of
13 irrigation withdrawals from shallow aquifers during the dry season is at least comparable to that
14 of annual vertical recharge (Harvey et al., 2002; 2006). Irrigation could therefore significantly
15 affect the distribution of shallow groundwater properties in the study area.

16
17 Is the spatial relation between groundwater age profiles and surface permeability inferred from
18 EM conductivity consistent with plausible flow lines for recharge? Based on the simple
19 calculation of the lateral advection of recharged water, in areas where shallow groundwater is
20 young should roughly correspond to areas where surface permeability inferred from EM
21 conductivity is low (Fig. 9, Table 1). In contrast, in areas where surface permeability is low,
22 shallow groundwater that is older in age recharged at far distant areas (Fig. 9). Therefore, the
23 connection between groundwater age and surface permeability might be more complex because
24 of variable flow paths leading from more distant recharge areas. This might explain the weak
25 relationship between surface EM conductivity and groundwater As concentrations in wells (Fig.
26 10a). This might also help to elucidate the considerable scatter of the relationship between EM
27 conductivity and As-depth gradients that remains even after properties are averaged within a
28 radius of 50 m (Fig. 10c).

29
30 In a complementary study conducted in the same area, Weinman et al. (in press) have shown that
31 low-EM conductivity areas of Araihasar are typically associated with high energy, sandy
32 depositional environments such as relict channel levees and bars, on the basis of elevation
33 surveys and grain-size analysis of a considerable number of auger cores. In contrast, the geology
34 of several of the very high-EM conductivity areas was interpreted as an indication of abandoned
35 channels filled with fine-grained sediment. On the basis of these findings, the variability of
36 recharge of shallow aquifers in Araihasar can therefore also be related the depositional history of
37 fluvio-deltaic environments.

38 39 **5.5 Implications for groundwater As**

40 Stute et al. (2007) showed that As concentrations in shallow aquifers of Araihasar increase
41 roughly linearly with the age of the groundwater at a rate of ~20 ug/L yr⁻¹. Dissolved As
42 concentrations are typically well below 50 ug/L in Bangladesh pond and river water and, we can
43 safely assume, also in precipitation (BGS/DPHE, 2001). Vertical recharge from surface water
44 bodies and precipitation would therefore tend to dilute any As that is released from the sediment
45 at depth in reducing aquifers. Recharge with precipitation or surface water also supplies
46 oxidants in the form of dissolved oxygen and nitrate that inhibit the reductive dissolution of Fe

1 oxyhydroxides (BGS/DPHE, 2001; Horneman et al., 2004). Both dilution and inhibition of
2 reductive dissolution are consistent with the observation that dissolved As concentrations in the
3 shallowest aquifers of Araihasar decrease as the rate of local recharge increases.

4
5 Water levels in the confluence of the Old Brahmaputra River are rarely higher than in shallow
6 aquifers of the area (Fig. 5). Lateral flow across river banks may also be inhibited by fine-
7 grained river bottom and overbank deposits, suggesting that rivers may be a relatively
8 unimportant source of recharge in Araihasar. Perhaps for that reason also, the majority of
9 shallow tubewells along the banks of the Old Brahmaputra River and its confluence are elevated in
10 As (Fig. 1a). The situation may be different in an area such as Munshiganj where there is less
11 opportunity for vertical recharge because of the presence of an uninterrupted clay layer at the
12 surface (Harvey et al., 2006).

13
14 It remains unclear to what extent the relationship between surface permeability, vertical recharge,
15 and As concentrations reflects a steady source within the sediment that is diluted by groundwater
16 flow or the flushing of shallow aquifers of their initial As content over time (Radloff et al., 2007;
17 Stute et al., 2007; van Geen et al., in press). Such uncertainties make it difficult to speculate on
18 the impact that the onset of irrigation pumping in recent decades may have had on the
19 distribution of As in shallow groundwater. If the onset of irrigation pumping enhanced vertical
20 recharge, the weight of the evidence would suggest that at least in Araihasar this resulted in
21 lower rather than higher As concentrations in shallow aquifers (Harvey et al., 2002; 2006; Klump
22 et al., 2006; Stute et al., 2007).

23 24 **6. Conclusion**

25 The geophysical data presented in this study reveal a relationship between the permeability of
26 surface soils and the distribution of both groundwater ages and As concentrations in shallow
27 aquifers of Araihasar. The observations underline the importance of vertical recharge of
28 precipitation, monsoonal flooding, and irrigation seepage water for setting the properties of
29 shallow groundwater including As. Hydrological processes must therefore be considered to
30 unravel widespread but spatially variable As enrichments in South Asian deltaic aquifers, in
31 conjunction with the still poorly understood biogeochemical processes that lead to the release of
32 As to groundwater.

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Figure Captions

Figure 1 (a) Distribution of As concentrations in 4700 hand-pumped wells <22 m (75 ft) deep within the study area in Araihaazar shown as color-coded circles. The locations of 18,530 EM conductivity measurements are shown as gray lines. Open black circles show the location of mechanized irrigation wells. (b) Distribution of EM conductivity in the fields of Araihaazar shown as color coded squares. In this map, the location of shallow private wells is shown by grey circles. Blue crosses indicate the location of soil profiles collected with a hand-auger. In addition, multilevel wells sites are shown as yellow circles and the river water level station is a yellow star. Open yellow circles identify the subset of 614 wells with at least one EM conductivity measurement with a radius of 50 m.

Figure 2 Distribution of As as a function of depth in nearly 4700 wells of Araihaazar <22 m (75ft) deep. The solid line corresponds to the relation obtained by least squares regression.

Figure 3 Histogram of EM conductivities measured in the Araihaazar study area.

Figure 4 Soil properties measured in cores collected with an auger at 14 locations: (a) electrical conductivity measured in soil slurries and (b) clay fraction. Profiles are color-coded in three groups on the basis of EM conductivity measured at the surface: (3-10 mS/m - green), medium (11-15 mS/m - blue), and high (16-35 mS/m - red).

Figure 5 Box plot showing hydraulic head differences of shallow (<15 m) wells at well nests relative to the water level of the confluence of the Old Brahmaputra River measured from a concrete bridge (Fig. 1b) over two and half years. Positive values correspond to a hydraulic gradient towards the river. Monthly measurements are averaged (a) over three periods: pre-monsoon (Feb-May), monsoon (Jun-Sept), and post-monsoon (Oct-Jan) and (b) over the whole 2004-2006 period of measurements. The black line in the middle of each box indicates the median; the dotted line corresponds to the average. The top side of the box indicates the third quartile and the bottom side the first quartile. The whiskers at either end of the box correspond to values within the fence (± 1.5 times the interquartile range (IQR)). Solid black circles indicate moderate outliers (outside the $1.5 \times$ IQR range).

Figure 6 Regression of (a) slurry conductivity and (b) clay fraction integrated to a depth of 200 cm (as described in the text) as a function of EM conductivity for 14 soil profiles collected in the Araihaazar study area.

Figure 7 Variogram for EM conductivity at an active lag of 1000 m and an active step of 10 m was constructed using GS+ ® v5.0 (Gamma Design, 2000). Small open circles show the experimental variogram; the solid line indicates the theoretical variogram.

Figure 8 Comparison of the distribution of As in shallow wells of Araihaazar with a contour map of EM conductivity (axis labels in decimal degrees). The EM conductivity surface was mapped using ordinary block kriging with a maximum distance of extrapolation of 100 m. Arrows point to the four well nests where recharge rates (numbers in m/y) were calculated from ^3H - ^3He ages (Stute et al., 2007).

Figure 9 Diagrams showing the range of distances traveled by groundwater after being recharged to reach the pumping wells assuming the wells are 15m deep (vertical scale is exaggerated). The calculation of distances is discussed in the text and is shown in the Table 1.

Figure 10 Comparison of (a) As content of 614 shallow wells as a function of the closest EM conductivity measurement obtained within a distance of 50 m. (b) As in 504 shallow wells as a function of average EM conductivity consisting of at least 5 measurements within a radius of 50 m. (c) As-depth gradient as a function of average EM conductivities for 145 circles of 50 m radius wells encompassing at least 5 EM conductivity and at least 5 household wells from which the local depth gradient for As could be calculated by least squares regression with non-negativity constraint on the intercept. Horizontal error bars show the standard deviation of at least 5 EM conductivity measurements. Vertical error bars show the standard error in the estimate of the slope of As versus depth by regression for at least 5 wells.

Figure 11 Depth profiles of As at the five wells nests (black circles), in order of increasing nearby EM conductivity. Open circles show the depth distribution of As in household wells located within 50 m of each well nest. Also shown are least squares regression lines calculated by imposing a non-negativity constraint on the intercept.

Figure 12 (a) Comparison of average EM conductivities and As-depth gradients calculated from multilevel well sites (black circles) and household wells (open circles) within 50 m of each well nest. Error bars indicate the standard deviation of EM conductivities and the uncertainty in the As-depth slope (1-sigma), respectively. (b) Comparison of average EM conductivities and recharge rates estimated at the five well nests by Stute et al. (2007).

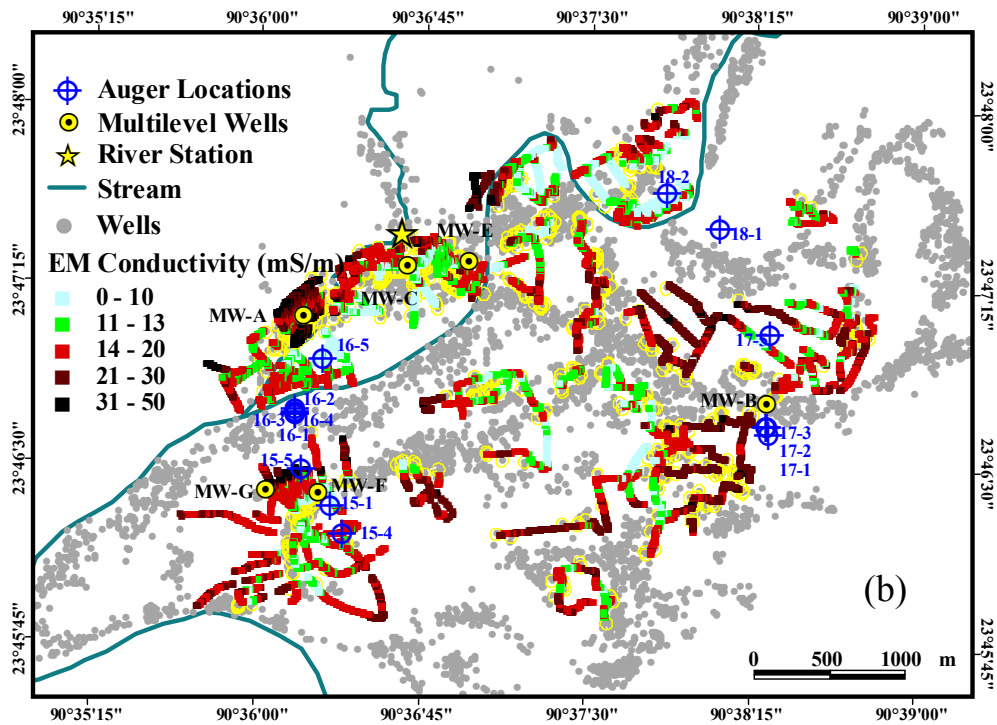
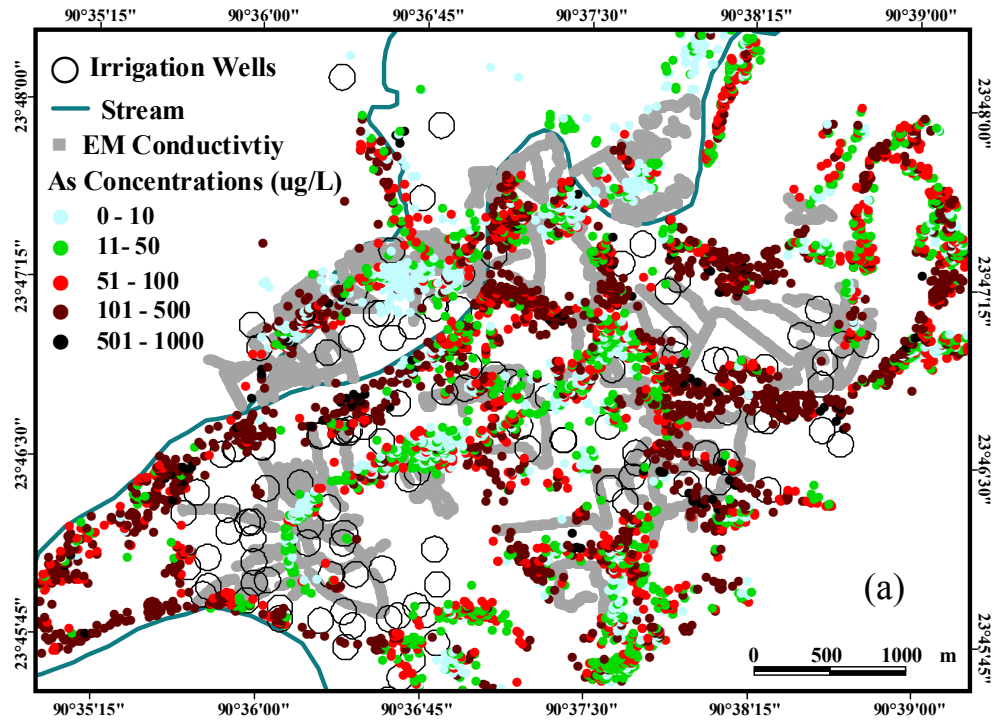


Figure 1

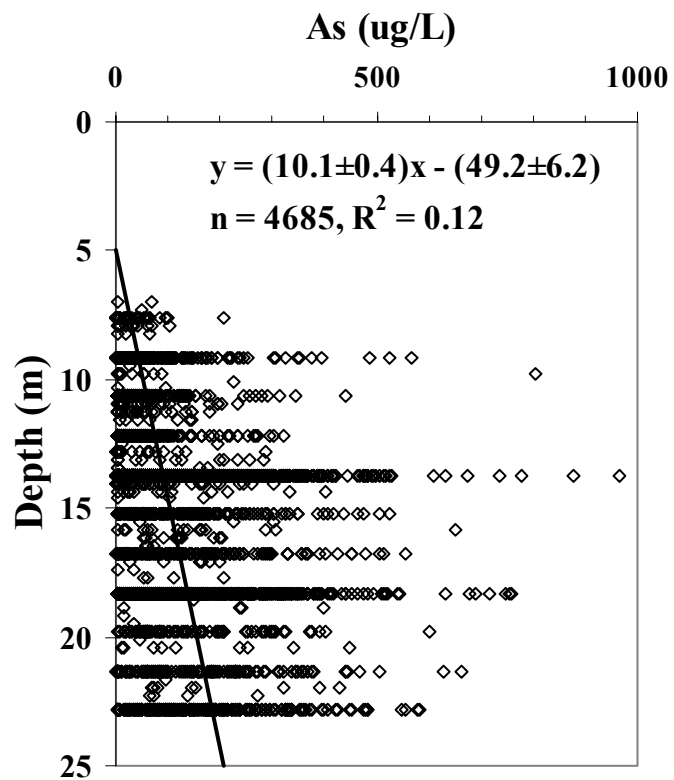


Figure 2

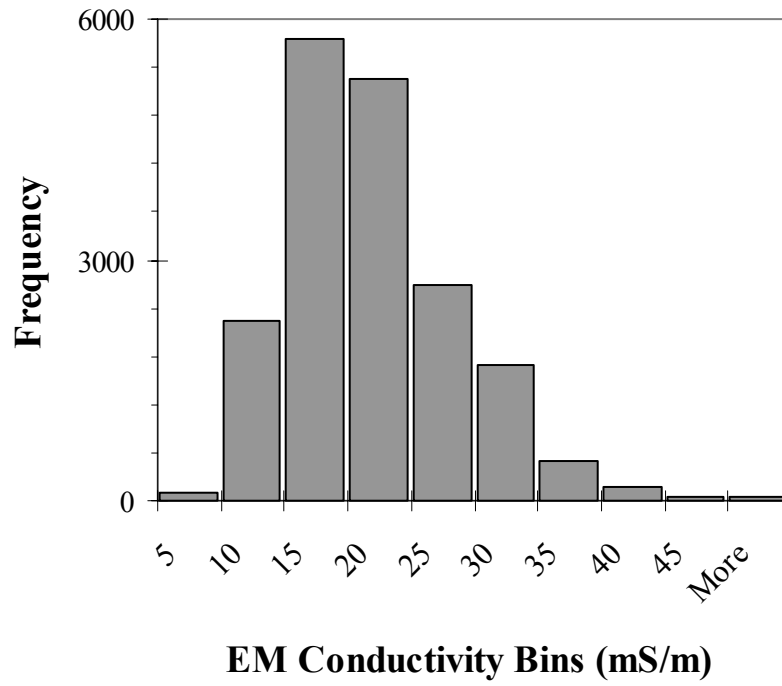


Figure 3

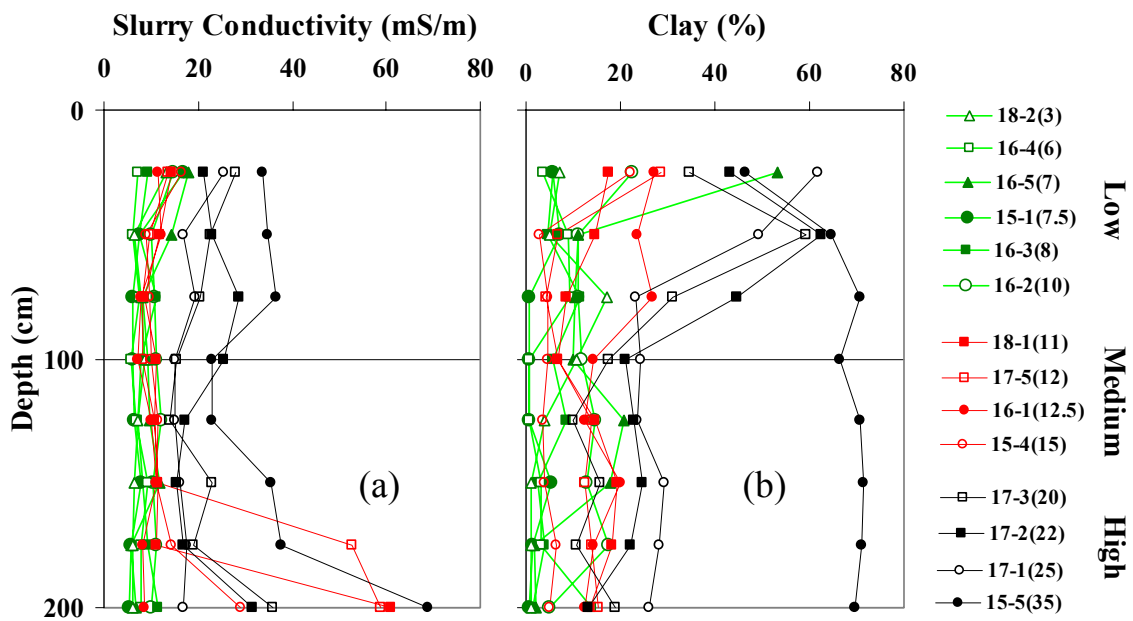


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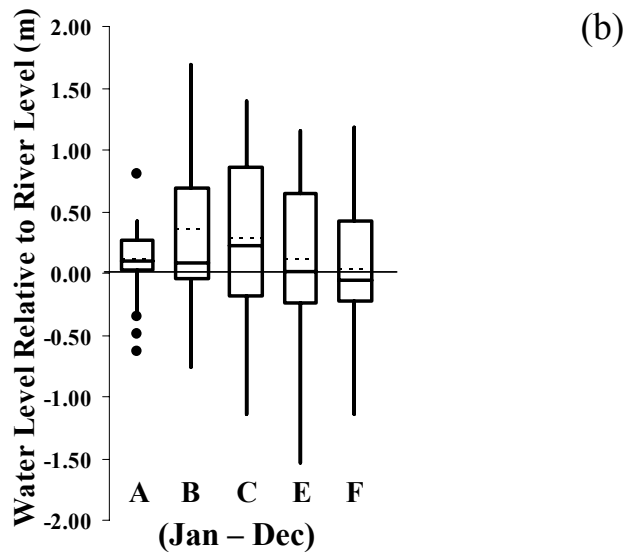
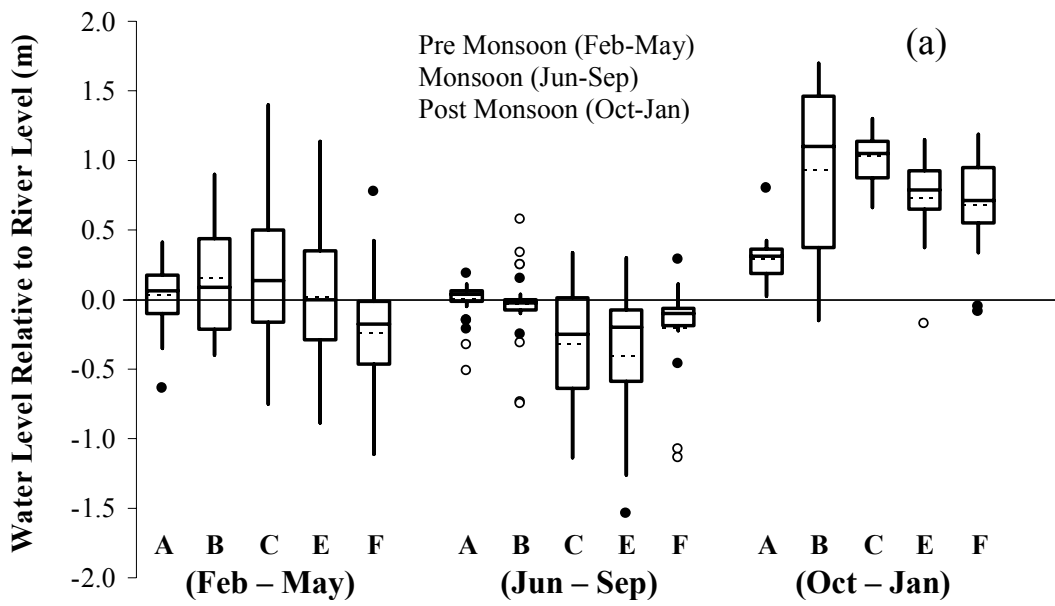


Figure 5

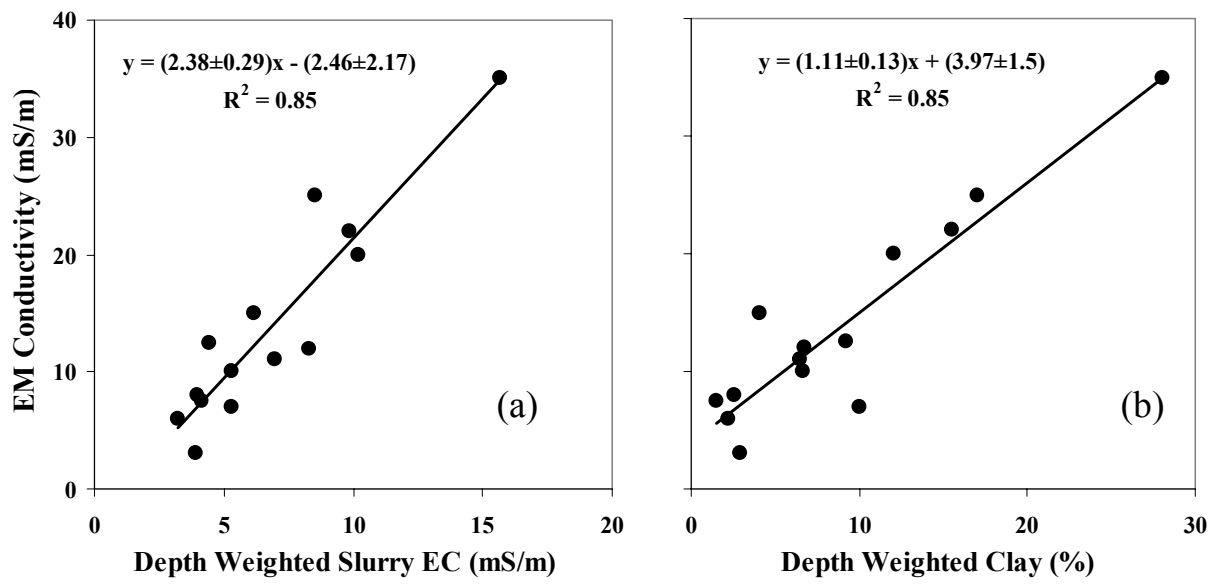


Figure 6

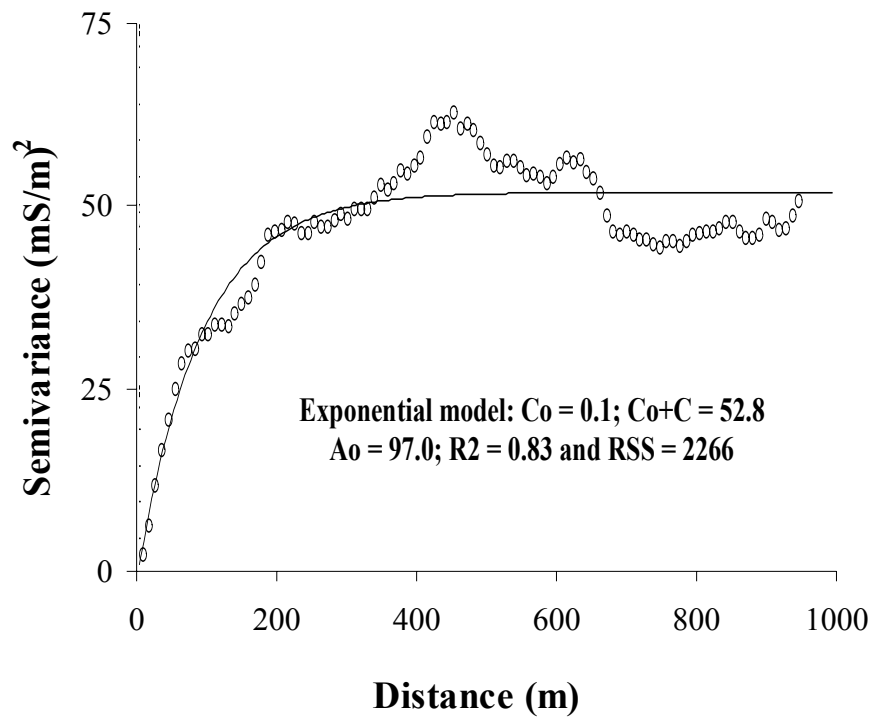


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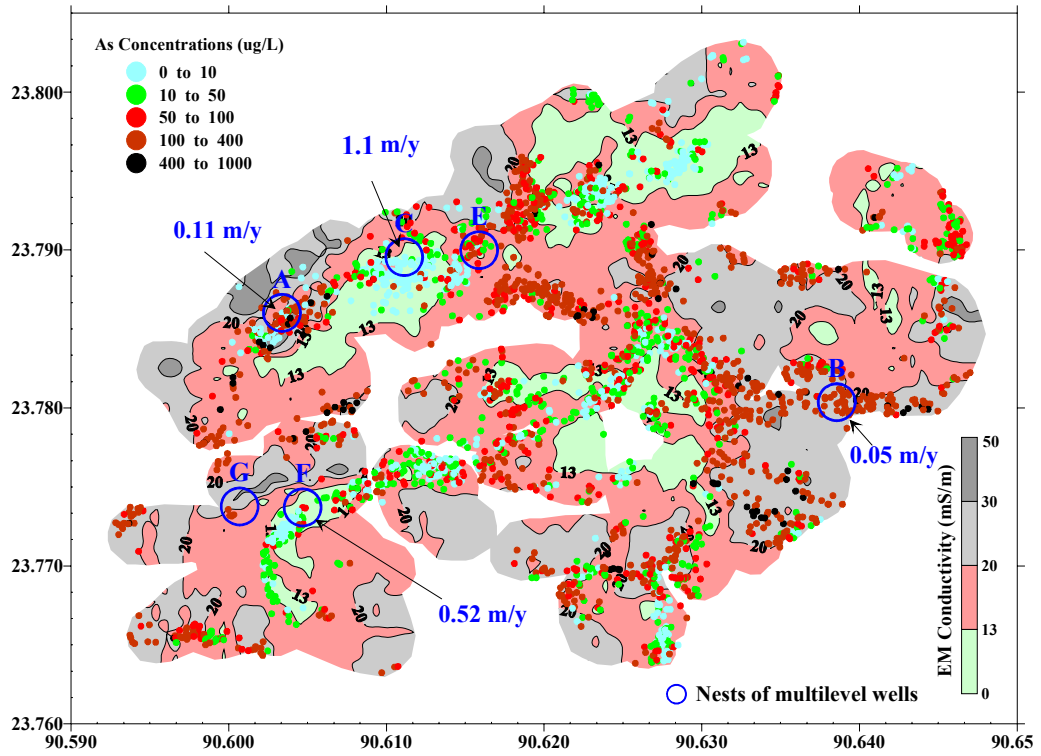


Figure 8

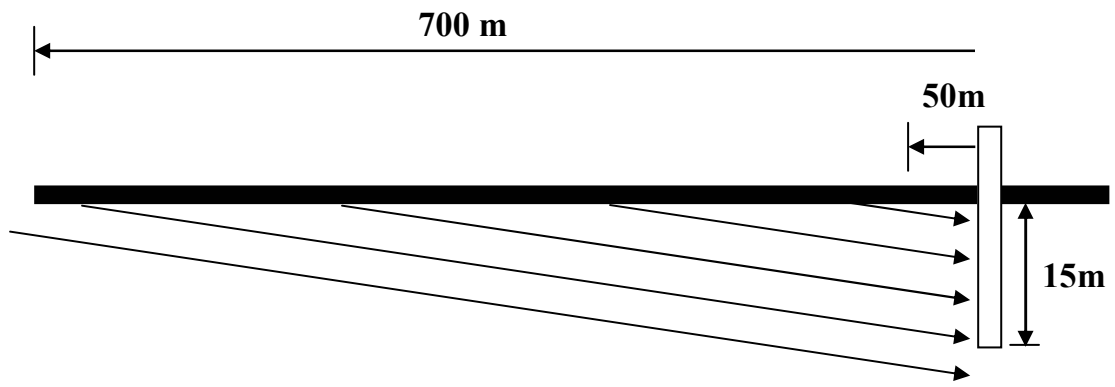


Figure 9

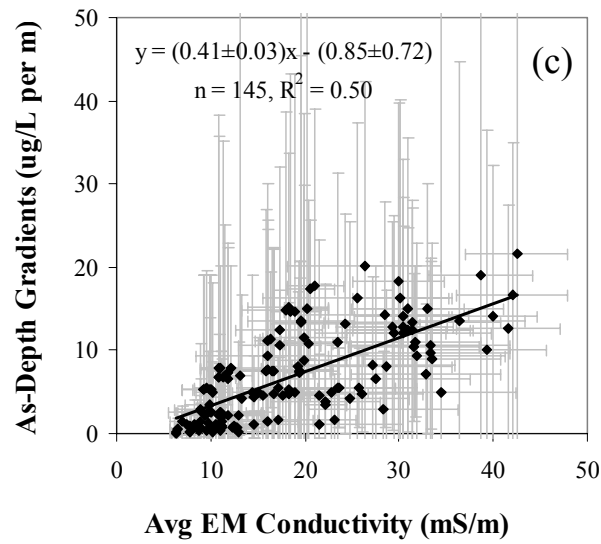
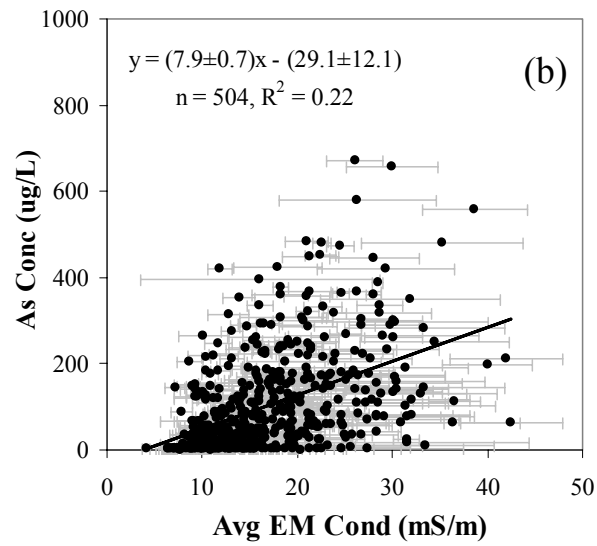
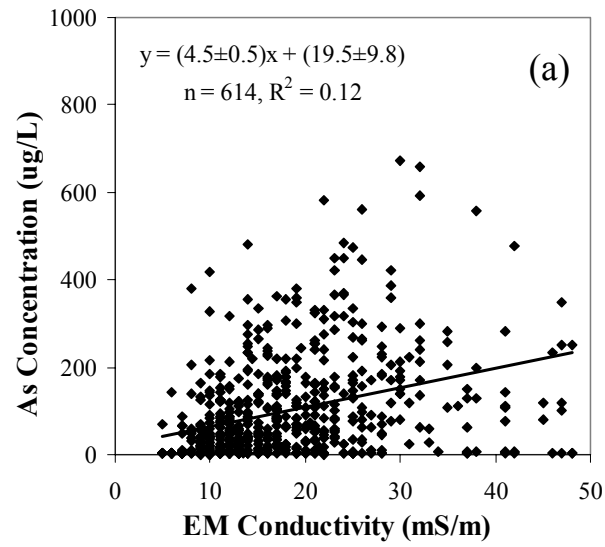


Figure 10

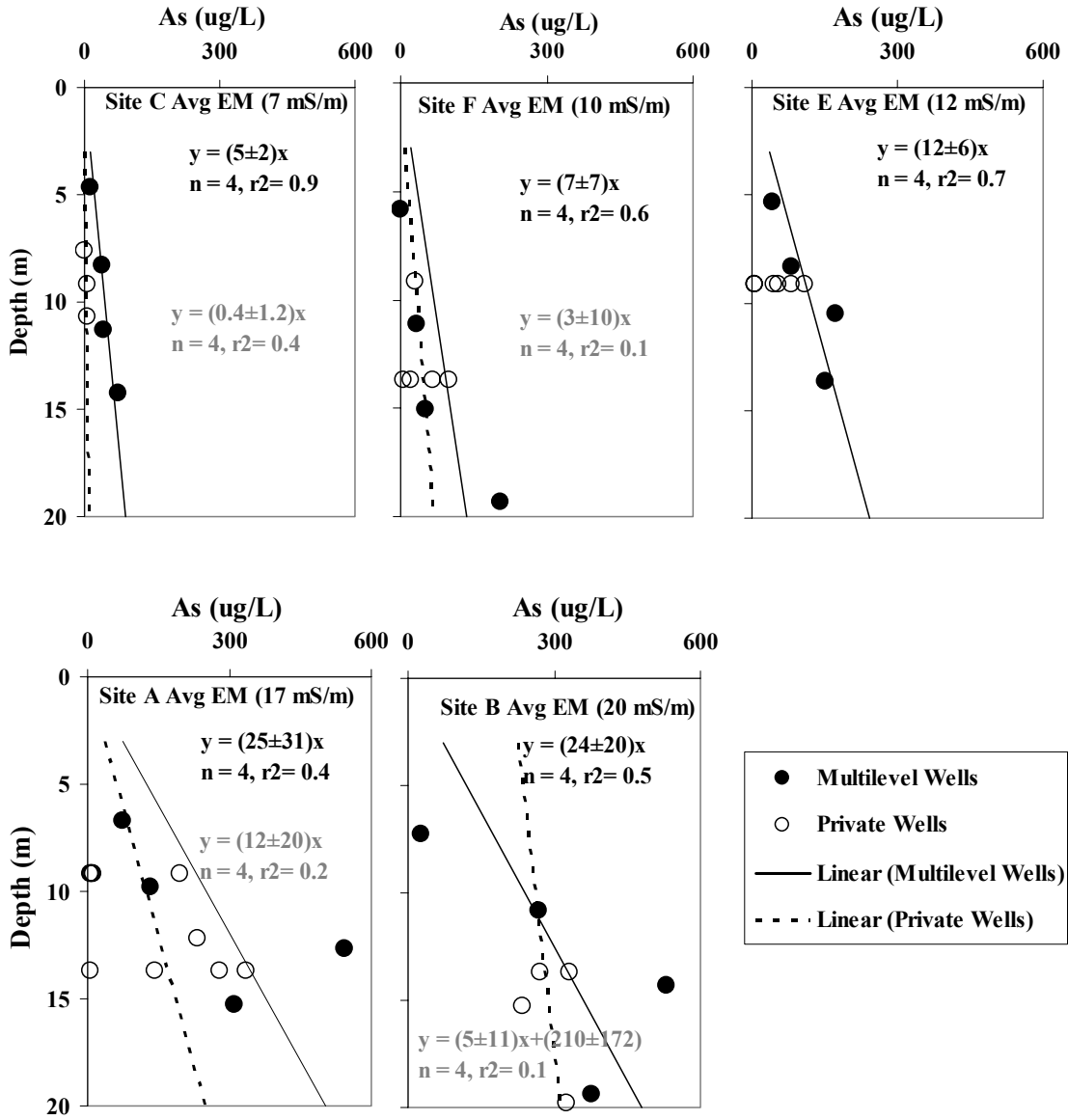


Figure 11

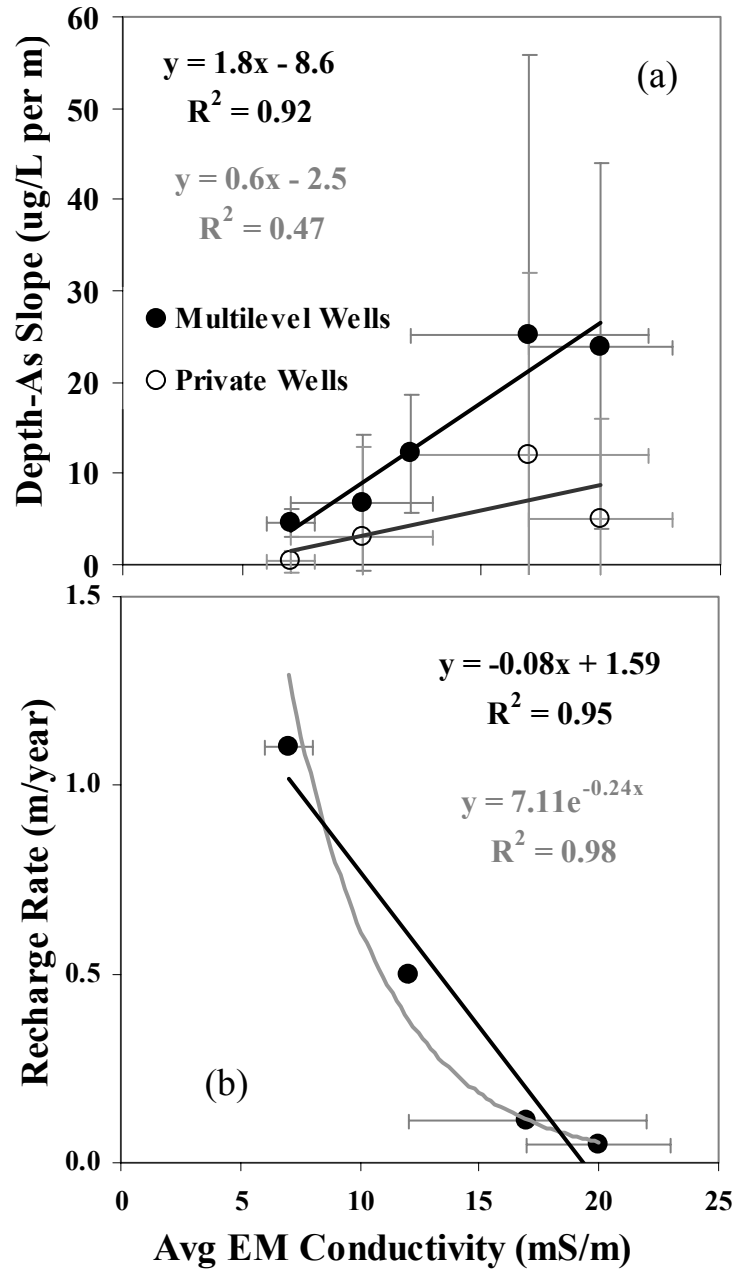


Figure 12

Table 1: Estimate of distance traveled ($x = r*z$) to reach a depth of 15m at each of the multilevel sites based on local hydraulic gradients and assuming a horizontal permeability 10-fold greater than vertical permeability. Distance to discharge at sites A and C is the actual distance to the Old Brahmaputra River and an estimate based on local topography at sites E and F. Horizontal head gradients are from annual averages in Fig. 5b and vertical head gradients are from Stute et al. (2007).

Site	Discharge Distance (m)	Horizontal Gradient ($i_x = dh/dx$)	Vertical Gradient ($i_z = dh/dz$)	Horizontal Flow/Vertical Flow ($r = 10*i_x/i_z$)	Recharge Distance (x) (m)
A	100	11.0E-04	6.7E-04	16.4	250
C	300	9.7E-04	3.3E-04	29.5	450
E	150	7.3E-04	1.7E-04	46.5	700
F	150	2.0E-04	4.3E-04	4.6	50

Table 2a: Regression analysis of As concentrations as a function of EM conductivity ($As = a \cdot EM \text{ Conductivity} + C$) for pairs of data separated by various distances. The relationships are all highly significant ($p < 0.001$)

Distance (m)	a	C	R ²	n
50	4.5±0.5	19.5±9.8	0.12	614
100	5.0±0.4	12.7±6.5	0.11	1555
200	5.2±0.3	13.9±5.5	0.10	2823
500	5.6±0.3	11.1±5.1	0.10	4002

Table 2b: Regression analysis of As concentrations as a function of average EM conductivity ($As = a \cdot \text{Avg EM Conductivity} + C$) for different minimum number of EM conductivity measurements within a radius of 50 m of each well. The relationships are all highly significant ($p < 0.001$)

Minimum EM Readings	a	C	R ²	n
1	5.7±0.6	-0.4±10.8	0.14	614
2	6.2±0.6	-4.1±11.2	0.15	587
5	7.7±0.7	-29.1±12.1	0.22	504
10	8.5±0.8	-38.3±14.1	0.23	395
20	8.3±1.0	-34.5±18.5	0.22	246

Table 3: Regression analysis of As-depth gradients as a function of average EM conductivity (As-Depth Gradients = $a \cdot \text{Avg EM Conductivity} + C$) for different minimum number of EM conductivity measurements and wells within a radius of 50 m of each well. As-Depth gradients were calculated using non-negativity constrained least squares method. The relationships are all highly significant ($p < 0.001$)

Min # of Wells	Min # of EM	a	C	R²	n
3	1	0.43±0.03	-0.70±0.69	0.37	276
3	5	0.42±0.04	-0.43±0.73	0.37	233
3	10	0.40±0.04	0.07±0.88	0.32	180
3	20	0.39±0.06	0.14±1.12	0.29	122
5	1	0.43±0.03	-1.19±0.60	0.53	180
5	5	0.41±0.03	-0.85±0.72	0.50	145
5	10	0.40±0.04	-0.33±0.91	0.44	109
5	20	0.41±0.06	-0.74±1.17	0.41	79
3	1	0.43±0.03	-0.70±0.69	0.37	276
5	1	0.43±0.03	-1.19±0.60	0.53	180
10	1	0.44±0.03	-2.05±0.69	0.76	63