Impact of local recharge on arsenic concentrations in shallow aquifers inferred from the electromagnetic conductivity of soils in Araihazar, Bangladesh

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Received 27 February 2007; revised 10 January 2008; accepted 25 February 2008; published 26 July 2008.

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

Citation: Aziz, Z., et al. (2008), Impact of local recharge on arsenic concentrations in shallow aquifers inferred from the electromagnetic conductivity of soils in Araihazar, Bangladesh, *Water Resour. Res.*, *44*, W07416, doi:10.1029/2007WR006000.

1. Introduction

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

[3] In an attempt to decompose the groundwater As problem into more tractable questions, the present study focuses on the processes that regulate As concentrations in shallow aquifers of relatively recent ± <10 ka (i.e. Holocene) age. The field observations extend over a 25 km^2 area of Araihazar upazila where the distribution of As in the groundwater has been mapped at unprecedented resolution by sampling several thousand tubewells [*van Geen et al.*, 2003b, Figure 1a]. When considering only shallow wells, defined here as wells <22 m (75 ft) deep (metric and imperial units of depth are listed because the latter are more widely used in Bangladesh), Araihazar marks a transition zone between the coastal region to the south, where very few wells meet the 50 μg/L Bangladesh

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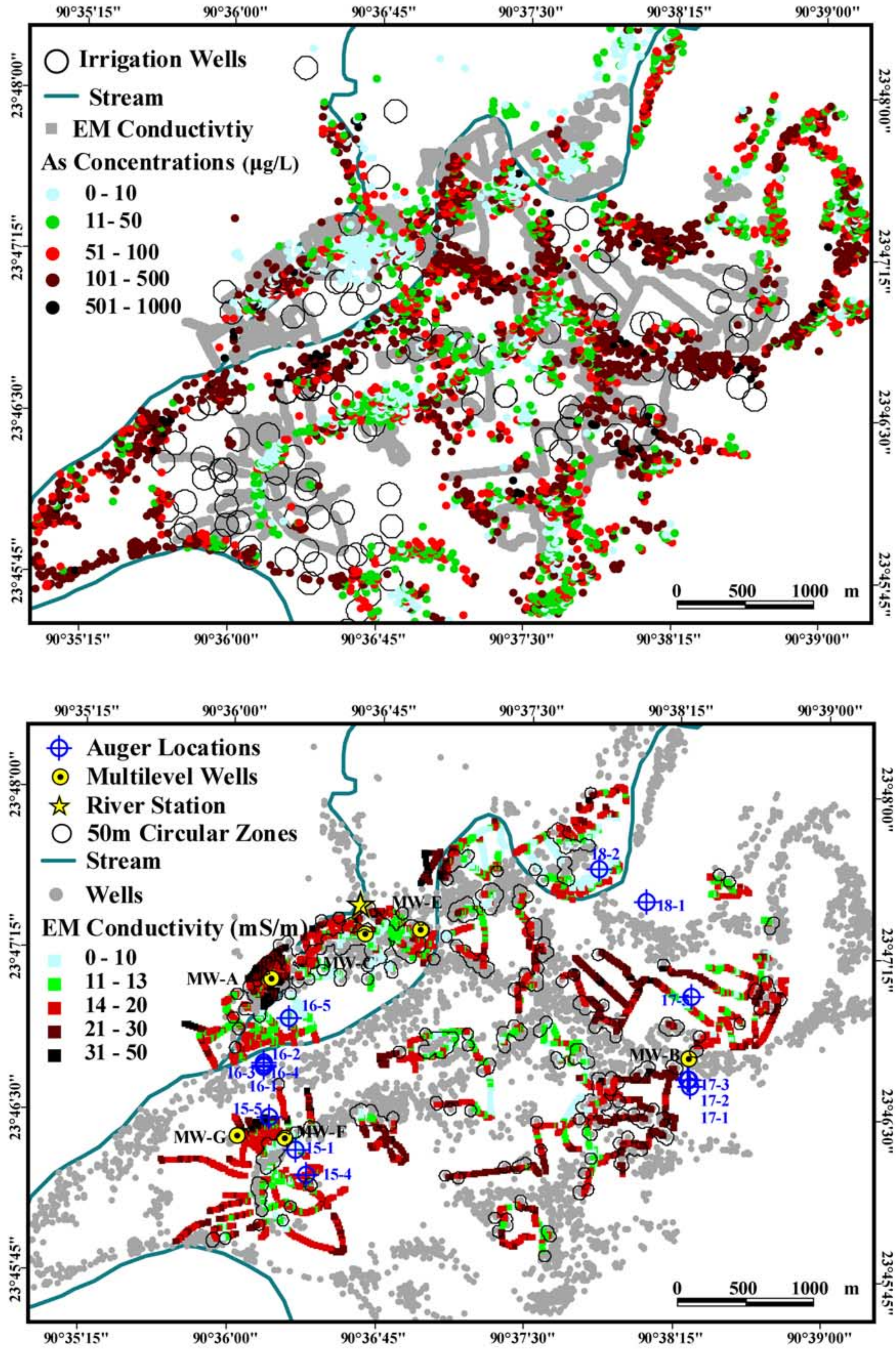
Irrigation Wells
Stream
EM Conductivtiy
As Concentrations (µg/L)
0 - 10
11- 50
51 - 100
101 - 500
501 - 1000
Auger Locations
Multilevel Wells
River Station
50m Circular Zones
Stream
Wells
EM Conductivity (mS/m)
0 - 10
11 - 13
14 - 20
21 - 30
31 - 50
MW-A
MW-B
MW-C
MW-E
MW-F
MW-G
0 500 1000 m

Figure 1

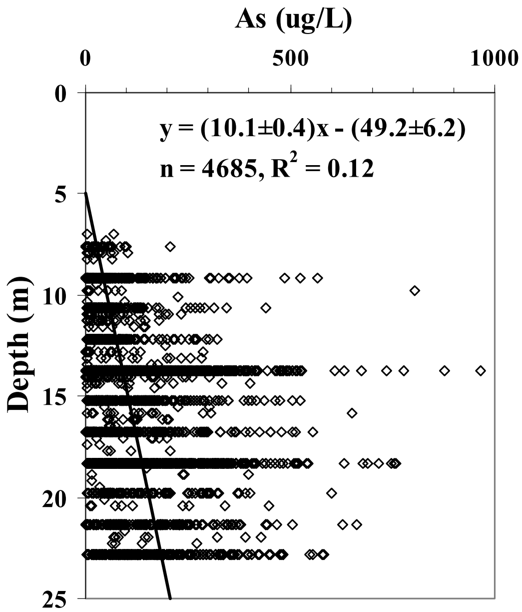

Figure 2. Distribution of As as a function of depth in nearly 4700 wells of Araihazar <22 m (75 ft) deep. The solid line corresponds to the relation obtained by least squares regression.

standard for As in the drinking water, and inland areas to the north where most shallow wells meet the 10 μg/L WHO guideline for drinking water As [*BGS/DPHE*, 2001]. Within Araihazar and other similarly affected regions of Bangladesh, average groundwater As concentrations in shallow aquifers generally increase with depth at any particular location [*BGS/DPHE*, 2001; *Harvey et al.*, 2002; *Horneman et al.*, 2004]. The relationship is highly scattered when extended over larger areas, however. Even within the confines of our 25 km^2 study area, there are numerous tubewells that are elevated in As and others that are low in As throughout the 8–22 m depth range (Figure 2). The present study is an attempt to shed some new light on the origin of this spatial variability by applying, to our knowledge for the first time in Bangladesh, an established and simple geophysical surveying technique, frequency domain electromagnetic induction, to relate spatial variations in the nature of surface soil properties to As concentrations in shallow groundwater.

[4] Frequency domain electromagnetic (EM) induction has been used in hydrology to map percolation of water in the vadose zone, to estimate the extent and internal structure of shallow aquifers, and to determine the extent of groundwater contamination [*McNeill*, 1980a, 1990; *Cook et al.*, 1989; *Pellerin*, 2002; *Pettersson and Nobes*, 2003]. EM conductivity has also helped classify soil types in a mid-Atlantic coastal plain [*Anderson-Cook et al.*, 2002]. A number of studies have shown that hand-held EM instruments deployed by walking through cultivated fields can also be used to determine the spatial variability of soil salinity [*McNeill*, 1986; *Hendrickx et al.*, 1992; *Lesch et al.*, 1995; *Vaughan et al.*, 1995; *Doolittle et al.*, 2001; *Triantafilis et al.*, 2002]. It has, however, typically been difficult to disentangle the direct contribution of fine-grained particles to EM conductivity from an indirect contribution due to the accumulation of salt by evaporation at the surface of relatively impermeable deposits [*Williams and Hoey*, 1987; *Cook et al.*, 1989, 1992; *Doolittle et al.*, 1994; *Johnson et al.*, 2001; *Triantafilis et al.*, 2002]. In addition to salt content and grain-size, some studies have shown that variations in soil moisture can influence the EM conductivity signal [e.g., *Waine et al.*, 2000] whereas, in a different setting, *Banton* [1997] observed little difference in EM response between wet and dry conditions.

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

2. Hydrogeological Setting

[6] Erosion of mountains surrounding the Bengal Basin has deposited thick layers of clay, silt, sand, and gravel across much of Bangladesh over the past 7 ka of the Holocene [*Morgan and McIntire*, 1959; *Umitsu*, 1993; *Goodbred and Kuehl*, 2000; *BGS and DPHE*, 2001; *Goodbred et al.*, 2003]. Our study area within Araihazar straddles a transitional region that extends from the uplifted Madhupur Tract to the northwest, where only surface soils contain recently deposited material, to the active floodplain of the Meghna River to the southeast where up to 150 m of sediment accumulated during the Holocene [*van Geen et al.*, 2003b; *Zheng et al.*, 2005]. The Old Brahmaputra River running through the study area today is a modest stream

Figure 1. (a) Distribution of As concentrations in 4700 hand-pumped wells <22 m (75 ft) deep within the study area in Araihazar 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 Araihazar 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 as a yellow star. Open black circles identify the subset of 614 wells with at least one EM conductivity measurement within a radius of 50 m.

that stops flowing during the dry season (Figures 1a and 1b), but there are indications of a larger river passing through the area that may have shifted its course in the 18th century along with the main channel of the Brahmaputra River [*Fergusson*, 1863; *Brammer*, 1996; *Weinman et al.*, 2008].

[7] Time series of river stages and groundwater levels as well as measurements of vertical hydraulic gradients at six multilevel well sites in Araihazar over 2 years show a seasonal pattern in groundwater levels that mimics the river stages, with different time lags during the rising and the falling periods [*Zheng et al.*, 2005; *Stute et al.*, 2007]. At the end of the dry season, there is a slight delay in the response of groundwater to rapidly rising water levels in rivers that is accompanied by an upward hydraulic head gradient, suggesting a lateral input of water from rivers to aquifers. There are also short periods at the beginning of the wet season when groundwater levels rise above the river and vertical hydraulic head differences point downward, probably due to recharge with precipitation. Whereas during the wet season (June–October), groundwater and river levels are comparable, the drop in groundwater levels considerably lags the decline in river levels into the dry season. Such seasonal differences indicate that recharge occurs rapidly over a larger area through bank filtration, flooding, and infiltration of rainwater, while discharge occurs at fewer locations such as local surface water bodies and by localized evapotranspiration. Groundwater flow during the dry season is also likely to be affected by irrigation pumping during the 2–4 month period when mechanized tubewells are used throughout the area to flood rice paddies with groundwater. Similar observations have been reported elsewhere in Bangladesh [*BGS/DPHE*, 2001], including a study area in Munshiganj, ~30 km southwest of Araihazar [*Harvey et al.*, 2002, 2006; *Klump et al.*, 2006]. In Munshiganj, shallow aquifers are semiconfined because of extensive surficial deposits of clay and fine clay. Whereas fine-grained surface deposits are observed in Araihazar, there are also large areas where sandy deposits extend to the surface and are not capped by an impermeable layer [*Horneman et al.*, 2004; *Stute et al.*, 2007; *Weinman et al.*, 2008]. Direct rainfall, flooding, and seepage from irrigated fields and surface water bodies such as ponds may therefore constitute a larger component of recharge in Araihazar compared to Munshiganj [*Harvey et al.*, 2006; *Stute et al.*, 2007].

3. Methods

3.1. EM Conductivity Survey

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

[9] Nearly 18,500 EM31 readings were collected at waist level in Araihazar, primarily during the dry months of January 2001, January 2002, and March–June 2002 (Figures 1a and 1b). Flooding precluded data collection during the wet season. At the beginning of each day, the instrument was calibrated following the standard procedure described in the operating manual [*Geonics Limited*, 1992]. The distance between sequential measurements along a particular transect ranged from 5 to 10 wide steps, i.e., ~4 to 8 m. The reproducibility of readings at a single location was generally within 0.5 mS/m. Each determination rounded to the nearest unit of mS/m was entered by hand in the field on a Compaq Pocket PC connected to a Global Positioning System (GPS) receiver using ESRI® ArcPad software. EM conductivity measured along the edge of flooded rice fields was typically ~1 mS/m higher than over adjacent dry areas. Coverage is limited to open areas to avoid interference from corrugated iron plates used in the villages for building and fences. EM data were also not collected within 10–20 m of overhead power lines.

3.2. Collection and Analysis of Soil Samples

[10] At 14 locations spanning the spectrum of EM conductivities measured in Araihazar (Figures 1a and 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 slurries 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 h and ranged from 0.5 % by weight in the case of sands to 30% in clays.

3.3. Hydraulic Head Measurement

[11] *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 (Figure 1b). These measurements were extended by a 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 biweekly 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

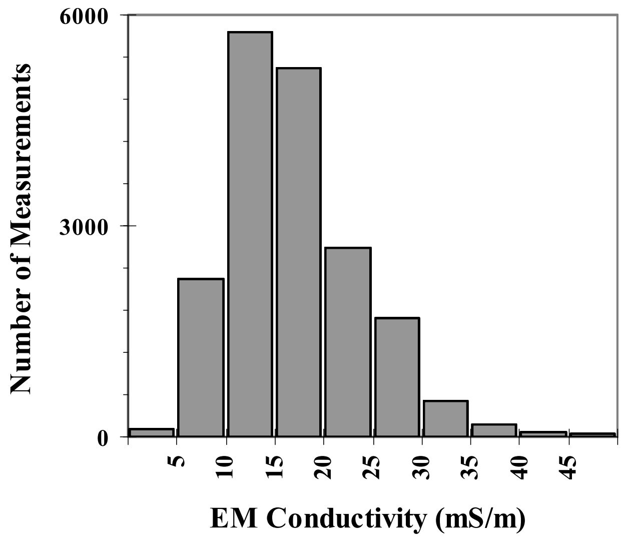

Figure 3. Histogram of EM conductivities measured in the Araihazar study area.

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

[12] EM conductivities measured in open areas of Araihazar 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 toward higher values, with a median of 17 mS/m and a variance of 48 mS/m (Figure 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 (Figure 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 μg/L (Figure 1a).

[13] 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 (Figure 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 (Figure 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.

4.2. Profiles of Soil Properties

4.2.1. Conductivity of Soil Slurries

[14] EM conductivities range from 3 to 35 mS/m at the 14 sites where auger cores were collected (Figure 1b). The conductivity of slurries of the underlying soil measured with a conductivity probe, referred to hereon as slurry conductivity, range from 5 to 60 mS/m. For ease of viewing, the profiles of slurry conductivity are subdivided into three groups. Slurry conductivities average 12 ± 12 mS/m (n = 112) and are essentially uniform with depth for the six profiles in the lowest category of EM conductivities (3–10 mS/m; Figure 4a). This is also the case for 1 out of 4 profiles

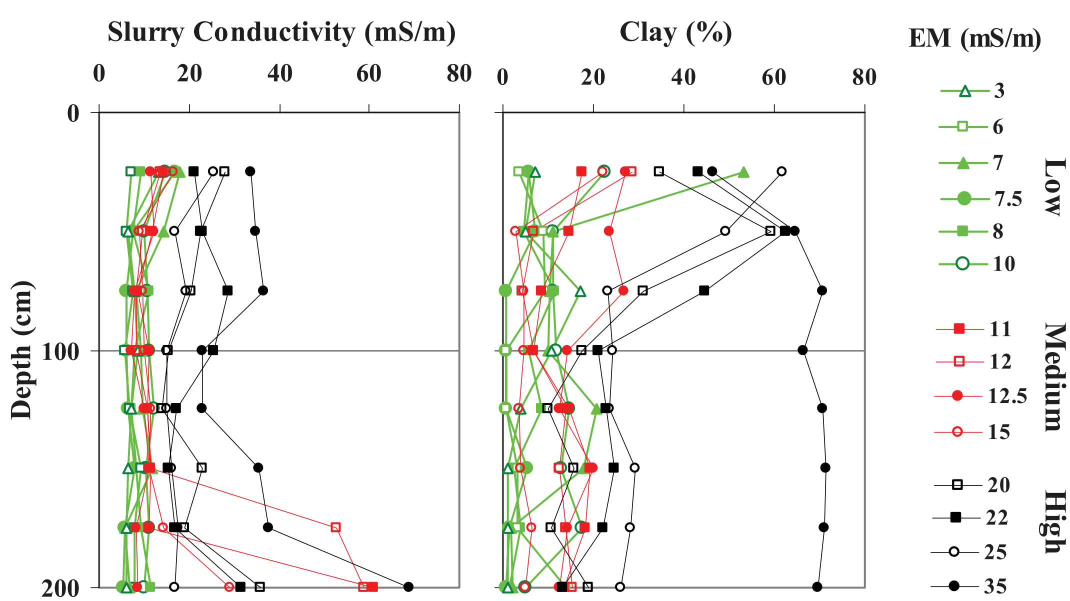

Figure 4. Soil properties measured in cores collected with an auger at 14 locations: (a) electrical conductivity measured in soil slurries and (b) clay fraction. Profiles are color-coded in three groups on the basis of EM conductivity measured at the surface: (3–10 mS/m, green), medium (11–15 mS/m, red),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

from sites in the intermediate EM conductivity range (11–15 mS/m; Figure 4a). The other three profiles in this category, however, show two- to six-fold increases in slurry conductivity in the deepest sample at 200 cm. A similar increase in conductivity at depth is observed for three out of six profiles in the highest category of EM conductivities (20–35 mS/m; Figure 4a). In this category, slurry conductivities in the upper 150 cm of the soil are also higher (14 ± 8 mS/m, n = 14) than for the two groups of sites showing lower EM conductivities.

4.2.2. Grain-Size

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

4.3. Water Levels in Araihazar

[16] Hydraulic heads are displayed as box plots of the difference in head between the shallowest (<15 m) well from five multilevel well sites and the river for the premonsoon (February–May), monsoon (June–September), and postmonsoon (October–January) period (Figure 5a). Positive head differences from each monitoring site indicate potential groundwater flow toward the river and vice versa. The average water level in the shallowest well from each site remains close to that of the river during the monsoon at three of the sites (sites A, B, and F). The difference in water levels is more variable at the two other sites (sites C and E) and on average below that of the confluent of the Old Brahmaputra River, suggesting more local control. Groundwater levels in shallow wells at four of the sites remain at or above that of the river during the four months preceding the monsoon (Figure 5a). The one exception is Site F where irrigation is particularly intense (Figure 1a) and average water levels in shallow aquifers remain below that of the river (Figure 5a). At all five sites, however, average hydraulic gradients over the entire 2-year period point toward the river (Figure 5b).

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

5. Discussion

5.1. EM Conductivity and Sediments Properties

[18] From first principles, *McNeill* [1980a] showed that the contribution from different depths to EM conductivity can be calculated as $S_A = S_1 [1 - R_V(Z_1)] + S_2[R_V(Z_1) - R_V(Z_2)] + \ldots + S_N R_V(Z_N)$, where $S_1 \ldots S_N$ are soil conductivities at depths $Z_1 \ldots Z_N$, and $R_V(Z) = \frac{4Z}{(4Z^2+1)^{3/2}}$ is the cumulative response function specifying the contribution from each depth, expressed as a fraction of the distance separating the emitting and receiving coils. We make here no attempt to calibrate the EM31 signal in any absolute sense, nor do we try to separate the contribution to EM conductivity of ions present in soil water and exchangeable ions at the solid/solution interface [*Rhoades*, 1981; *Rhoades and Corwin*, 1981; *Rhoades et al.*, 1989; *Cook et al.*, 1989]. Instead, we compute a depth-weighted slurry conductivity and a depth- weighted clay fraction from different soil layers by applying the formulation of *McNeill* [1980a] separately to the measured slurry conductivities and to the clay fractions at every auger site. Contributions from different depths were corrected according to the height (0.5m) of the instrument above the ground. EM conductivities measured over the 14 auger sites broadly increases with the integrated slurry conductivity (Figure 6a). EM conductivities also increase fairly systematically as a function of the depth-weighted proportion of clay in the soil (Figure 6b). Linear regression shows that depth-weighted slurry conductivity (r^2 = 0.85, n = 14) and depth-weighted clay fraction (r^2 = 0.85, n = 14) correlate with EM conductivities measured at the surface, and this despite the significant contribution to the EM signal of layers below 2 m depth.

[19] It is difficult to distinguish the contributions of the clay fraction and the conductivity of soil water to the EM signal. However, previous studies have shown that the controlling factor in some areas is clay content [*Dalgaard et al.*, 2001; *Durlesser*, 1999; *Hedley et al.*, 2004; *Triantafilis and Lesch*, 2005]. In a setting similar to Bangladesh, *Kitchen et al.* [1996] observed strongly reduced EM conductivity over sand deposits dating from the 1993 flood of the Missouri River compared to surrounding finer soils. *Doolittle et al.* [2002] also located subsurface sand blows in southeastern Missouri by EM conductivity. In Australia and in the US, EM conductivity surveys have been used

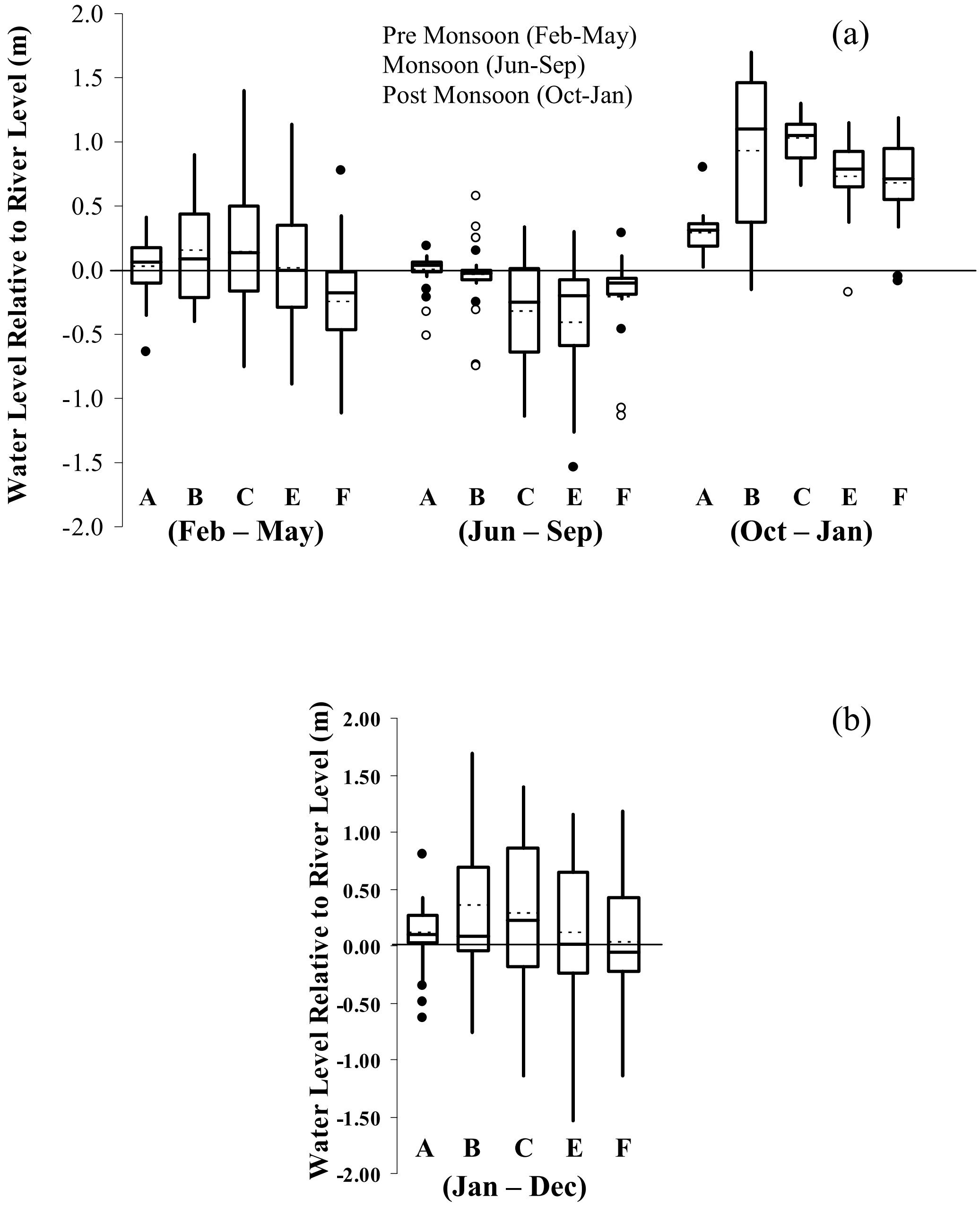

Figure 5. Box plot showing hydraulic head differences of shallow (<15 m) wells at well nests relative to the water level of the confluence of the Old Brahmaputra River measured from a concrete bridge (Figure 1b) over two and half years. Positive values correspond to a hydraulic gradient toward the river. Monthly measurements are averaged (a) over three periods: premonsoon (February–May), monsoon (June–September), and postmonsoon (October–January) 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 × IQR range).

to optimize rice production by identifying areas where irrigation water is lost to infiltration through sandy deposits [*Beecher et al.*, 2002; *Anderson-Cook et al.*, 2002]. What matters in the present context is that the auger cores show that the EM conductivity survey of Araihazar produced an extensive map of a property that is closely related to the clay content, and therefore the permeability, of surface soils.

5.2. Geostatistics of EM Conductivity

[20] The experimental variogram provides a convenient graphical representation of the continuity, or roughness, of spatial data [*Cressie*, 1993; *Robertson*, 2000; *Webster and Oliver*, 2001]. It is calculated using the equation $\gamma(h) = \frac{1}{2N(h)}\sum(Z_i - Z_{i+h})^2$ where γ(h) is semivariance

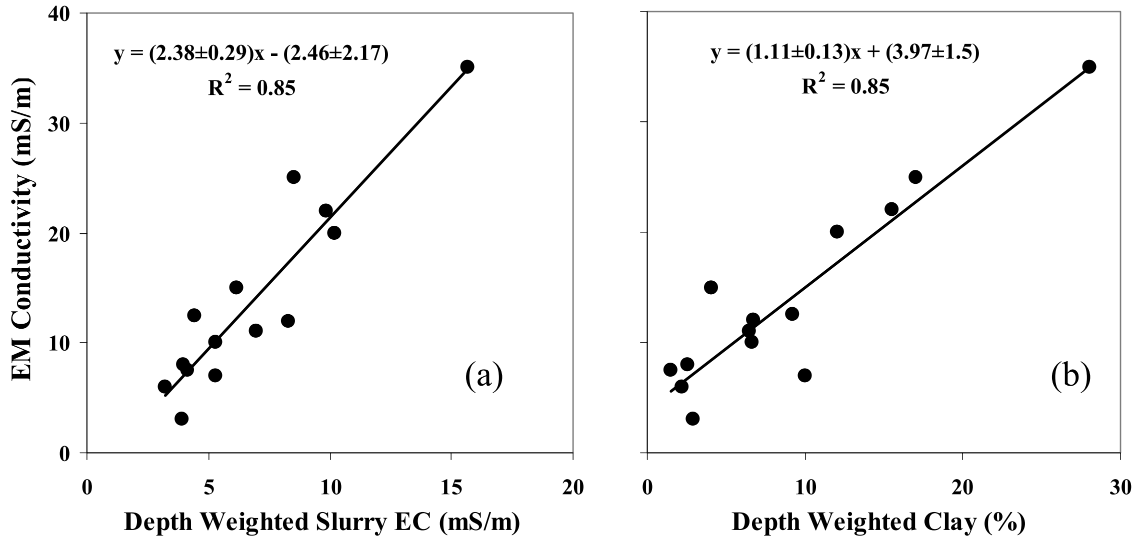

Figure 6. Regression of (a) slurry conductivity and (b) clay fraction integrated to a depth of 200 cm (as described in the text) as a function of EM conductivity for 14 soil profiles collected in the Araihazar study area.

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

5.3. Relation Between EM Conductivity and As in Groundwater

[21] The contour map of EM conductivity facilitates visual comparison with the distribution of As in shallow wells because the two data sets generally do not overlap (Figure 8). Merging of the two data sets shows, for instance, that interruptions of the diagonal swath of mostly low-As wells across the northwestern portion of the study area by clusters of high-As wells occur precisely in areas of elevated EM conductivity. Similarly, two clusters of generally low As concentrations to the south are closely delimited by areas of elevated EM conductivity. Overall, the average As content of shallow wells located within the 10 mS/m contour is 43 ± 67 μg/L (n = 606) whereas the average As content of wells located outside the same contour is 120 ± 123 μg/L (n = 2199). A different way to express the contrast between the two areas is that 73% of shallow wells located within the 10 mS/m contour contain <50 μg/L As, whereas this is the case for only

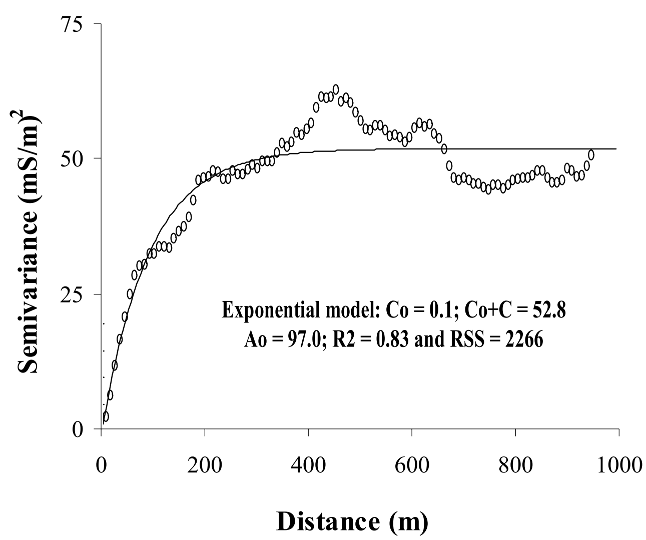

Figure 7. Variogram for EM conductivity at an active lag of 1000 m and an active step of 10 m was constructed using GS + ® v5.0 (Gamma Design, 2000). Small open circles show the experimental variogram; the solid line indicates the theoretical variogram.

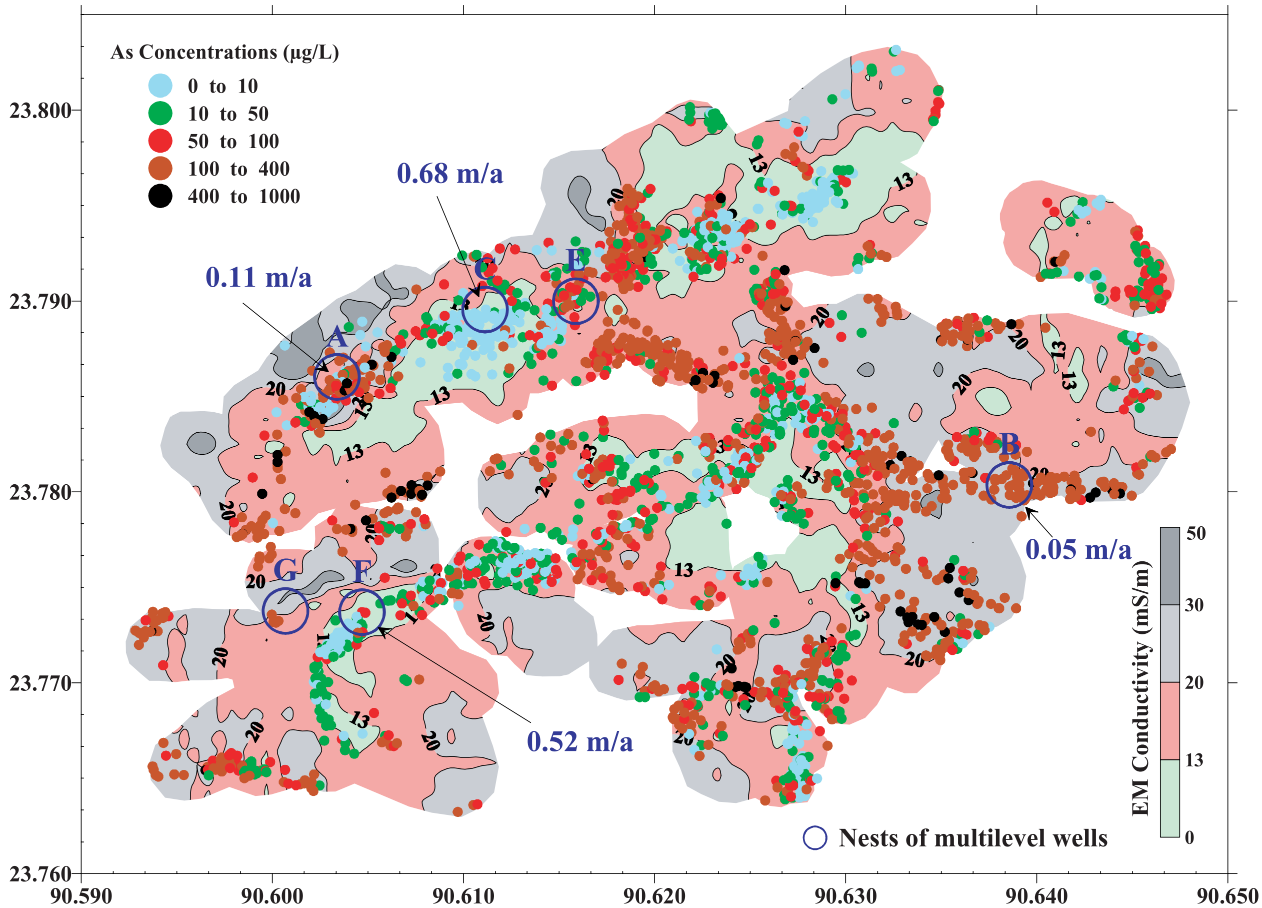

Figure 8. Comparison of the distribution of As in shallow wells of Araihazar with a contour map of EM conductivity (axis labels in decimal degrees). The EM conductivity surface was mapped using ordinary block kriging with a maximum distance of extrapolation of 100 m. Arrows point to the four well nests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

36% of wells outside the same contour. These observations suggest that shallow tubewells that meet the Bangladesh standard for groundwater of 50 μg/L As are unlikely to be found in areas where surface soils contain >10% clay (Figure 4).

[22] For a more quantitative comparison between groundwater As and EM conductivity data, we return to the actual EM data instead of extrapolated values by comparing the As content of each shallow well to the closest EM conductivity measurement. To determine the relevant spatial scales to consider, as determined by the ratio of horizontal to vertical flow, we estimate the distance over which recharged groundwater has to travel to reach a depth of 15 m (over two-thirds of shallow household wells considered in this study are up to 15 m deep). For lateral flow, we start from annually averaged relative hydraulic head differences between the multilevel sites and the Old Brahmputra River (Figure 5b). In the case of sites A and C, we use the actual distance of discharge to the river to calculate lateral gradients in average hydraulic head (100 and 300, respectively; Table 1). For sites of E and F, we estimate the shortest distance to a local discharge area to be on the order of 150 m rather than actual distances to the Old Brahmaputra River (Table 1). Vertical hydraulic gradients between the two shallowest wells at all four sites were previously reported by *Stute et al.* [2007]. Using the ratio between horizontal

Table 1. Estimate of Distance Traveled (x = r*z) to Reach a Depth of 15 m at Each of the Multilevel Sites Based on Local Hydraulic Gradients and Assuming a Horizontal Permeability 10-Fold Greater Than Vertical Permeability[a]

Site	Discharge Distance, m	Horizontal Gradient (ix = dh/dx)	Vertical Gradient (iz = dh/dz)	Horizontal Flow/Vertical Flow (r = 10*ix/iz)	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

[a]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 Figure 5b and vertical head gradients are from *Stute et al.* [2007].

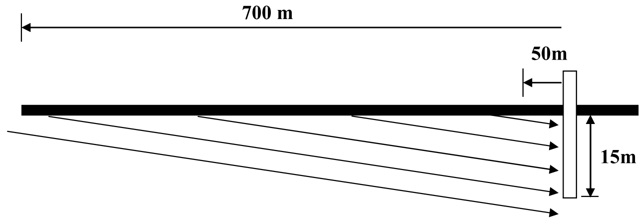

Figure 9. Diagrams showing the range of distances traveled by groundwater after being recharged to reach the pumping wells assuming the wells are 15 m deep (vertical scale is exaggerated). The calculation of distances is discussed in the text and is shown in Table 1.

and vertical gradients and assuming a vertical anisotropy (Kx/Kz) of 10 [*Freeze and Cherry*, 1979], distances over which recharged groundwater could have traveled to reach a depth of 15 m were then calculated. The results shows that groundwater from 15 m depth may have been advected laterally by 50 m at site F and by as much as 700 m at site E (Figure 9 and Table 1). This is just an order of magnitude estimate because the hydraulic gradient between wells nests and the river is not necessarily linear.

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

[24] Vertical profiles of As for nests of monitoring wells across the study area suggest that EM conductivities could instead be compared to the depth gradient of As concentrations in shallow aquifers rather than absolute As concentrations. Concentrations of As start from <50 μg/L in the shallowest monitoring well at each site and typically increase with depth, gradually at sites associated with low EM conductivities and more rapidly at high conductivity sites (Figure 11). Least squares regression of As as a function of

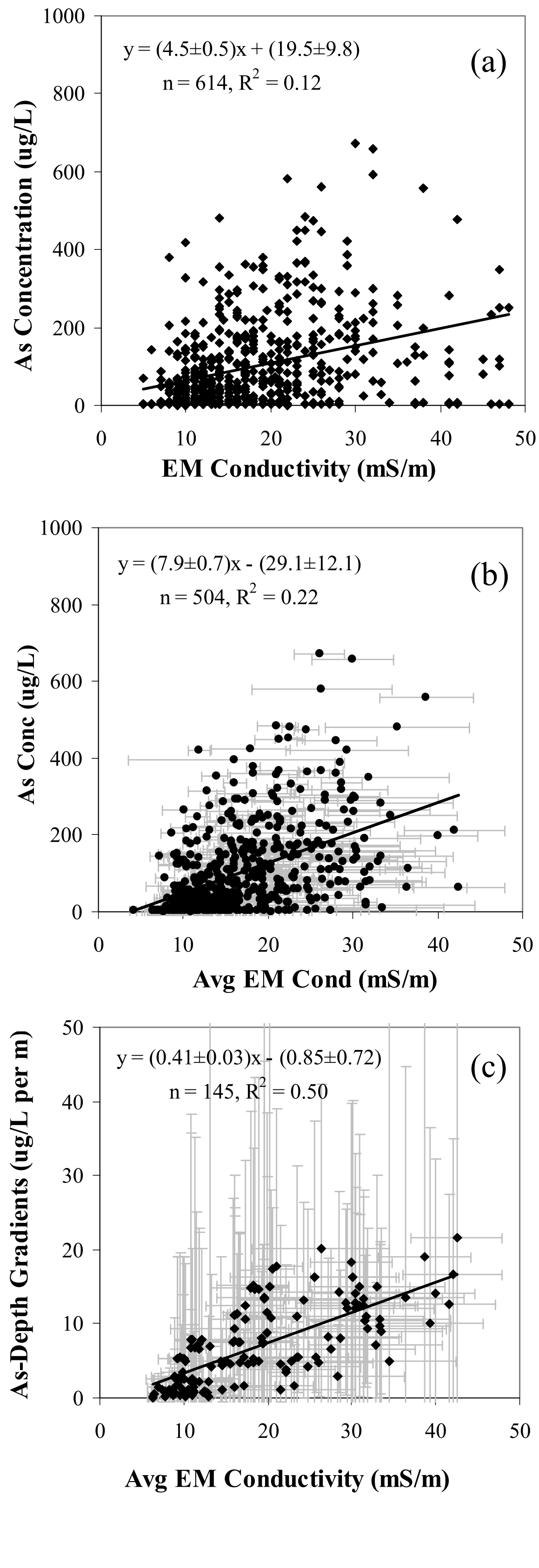

Figure 10. Comparison of (a) As content of 614 shallow wells as a function of the closest EM conductivity measurement obtained within a distance of 50 m. (b) As in 504 shallow wells as a function of average EM conductivity consisting of at least 5 measurements within a radius of 50 m. (c) As-depth gradient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

Table 2a. Regression Analysis of As Concentrations as a Function of EM Conductivity (As = a*EM Conductivity + C) for Pairs of Data Separated by Various Distances[a]

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

[a]The relationships are all highly significant ($p < 0.001$).

Table 2b. Regression Analysis of As Concentrations as a Function of Average EM Conductivity (As = a*Avg EM Conductivity + C) for Different Minimum Number of EM Conductivity Measurements Within a Radius of 50 m of Each Well[a]

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

[a]The relationships are all highly significant ($p < 0.001$).

depth, with a nonnegativity constraint imposed on the intercept, shows that the slope of this relationship increases systematically as a function of average EM conductivity within a radius of 50 m of each of these sites (Figure 12a). The slope of the As-depth relationship increases nearly five-fold across the 7–20 mS/m range in average EM conductivity at these five sites.

[25] When considering household wells located within 50 m of each of the five nests of the wells, the depth gradient in As concentrations is not as well defined, at least in part because household wells are typically deeper than the shallowest of the monitoring wells (Figure 11). The scatter of the depth trends in As is also considerably larger for household wells, possibly because of errors in reported well depths as well as lateral spatial heterogeneity. Despite

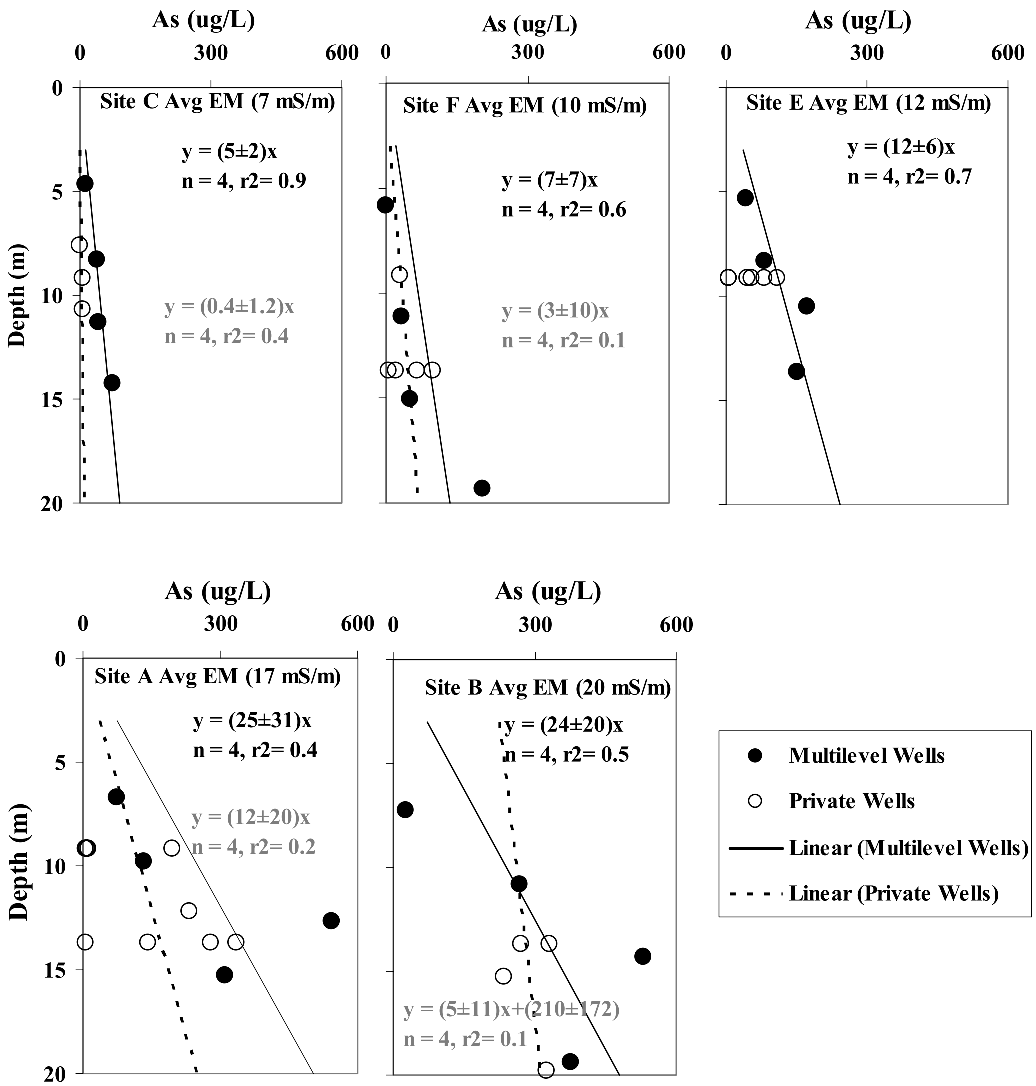

Figure 11. Depth profiles of As at the five well nests (black circles), in order of increasing nearby EM conductivity. Open circles show the depth distribution of As in household wells located within 50 m of each well nest. Also shown are least squares regression lines calculated by imposing a nonnegativity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

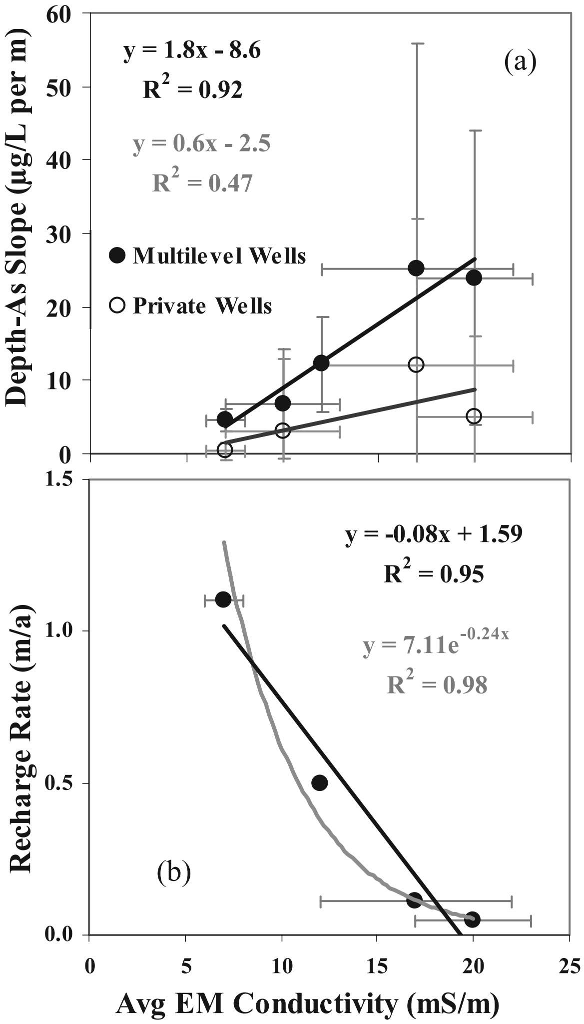

Figure 12. (a) Comparison of average EM conductivities and As-depth gradients calculated from multilevel well sites (black circles) and household wells (open circles) within 50 m of each well nest. Error bars indicate the standard deviation of EM conductivities and the uncertainty in the As-depth slope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

these limitations, the data show that the slope of the As-depth relationship based on household wells also generally increases as a function of EM conductivity (Figure 12a). It is unclear, however, why the depth gradients calculated from the household wells are a factor of three below the gradients calculated from the monitoring wells.

[26] The results from the area surrounding the five nests of wells justify a similar comparison of As depth gradients and EM conductivity within the larger data set. A total of 145 out of 614 circles of 50 m radius within the study area contain at least five household wells and at least 5 EM conductivity measurements (Figure 1b). Although there is considerable scatter, the relationship between EM conductivities and the As-depth gradient is highly significant ($p < 0.001$) and accounts for 50% of the variance (Figure 10c). Whereas the proportion of the variance that can be accounted for depends on the number of wells or EM measurements used for averaging, the relationship between As-depth gradients and EM conductivity itself is not very sensitive (Table 3). In the next two sections we examine why the rate of increase of As concentrations with depth in shallow aquifers of Araihazar might depend on the clay content of surface soil inferred from EM conductivity.

5.4. Relation Between EM Conductivity and Groundwater Age

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

[28] Similar conclusions were drawn from a combination of recharge estimates and electromagnetic measurements in southeastern Australia, albeit under considerably drier conditions than in Bangladesh [*Cook et al.*, 1992]. The same study predicts an exponential relationship between recharge and EM conductivity that may be consistent with our observations, even if a linear relationship could equally be inferred from our limited data (Figure 12b). The observations from Araihazar are more directly related to recent work conducted in the Himalayan foothills of India [*Israil et al.*, 2006]. In that study, the penetration of ^3H injected in surface soil by infiltration of precipitation and return seepage from irrigated fields was shown to increase with the

Table 3. Regression Analysis of As-Depth Gradients as a Function of Average EM Conductivity (As-Depth Gradients = a*Avg EM Conductivity + C) for Different Minimum Number of EM Conductivity Measurements and Wells Within a Radius of 50 m of Each Well[a]

Min # of Wells	Min # of EM	a	C	R^2	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

[a]As-Depth gradients were calculated using non-negativity constrained least squares method. The relationships are all highly significant ($p < 0.001$).

permeability of the soil, as determined by electrical resistivity measurements. Our observations from Araihazar provide further quantitative evidence that variations in surface permeability strongly influence the rate of local recharge of shallow fluvio-deltaic aquifers.

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

[30] Is the spatial relation between groundwater age profiles and surface permeability inferred from EM conductivity consistent with plausible flow lines for recharge? Based on the simple calculation of the lateral advection of recharged water, in areas where shallow groundwater is young should roughly correspond to areas where surface permeability inferred from EM conductivity is low (Figure 9 and Table 1). In contrast, in areas where surface permeability is low, shallow groundwater that is older in age recharged further away (Figure 9). Therefore the connection between groundwater age and surface permeability might be more complex because of variable flow paths leading from more distant recharge areas. This might explain the weak relationship between surface EM conductivity and groundwater As concentrations in wells (Figure 10a). This might also be the cause of the considerable scatter of the relationship between EM conductivity and As-depth gradients that remains even after properties are averaged within a radius of 50 m (Figure 10c).

[31] In a complementary study conducted in the same area, *Weinman et al.* [2008] have shown that low-EM conductivity areas of Araihazar are typically associated with high energy, sandy depositional environments such as relict channel levees and bars, on the basis of elevation surveys and grain-size analysis of a considerable number of auger cores. In contrast, the geology of several of the very high-EM conductivity areas was interpreted as an indication of abandoned channels filled with fine-grained sediment. On the basis of these findings, the variability of recharge of shallow aquifers in Araihazar can therefore also be related the depositional history of fluvio-deltaic environments.

5.5. Implications for Groundwater As

[32] *Stute et al.* [2007] showed that As concentrations in shallow aquifers of Araihazar increase roughly linearly with the age of the groundwater at a rate of ~20 μg/L per a. Dissolved As concentrations are typically well below 50 μg/L in Bangladesh pond and river water and, we can safely assume, also in precipitation [*BGS/DPHE*, 2001]. Vertical recharge from surface water bodies and precipitation would therefore tend to dilute any As that is released from the sediment at depth in reducing aquifers. Recharge with precipitation or surface water also supplies oxidants in the form of dissolved oxygen and nitrate that inhibit the reductive dissolution of Fe oxyhydroxides [*BGS/DPHE*, 2001; *Horneman et al.*, 2004]. Both dilution and inhibition of reductive dissolution are consistent with the observation that dissolved As concentrations in the shallowest aquifers of Araihazar decrease as the rate of local recharge increases.

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

[34] It remains unclear to what extent the relationship between surface permeability, vertical recharge, and As concentrations reflects a steady source within the sediment that is diluted by groundwater flow or the flushing of shallow aquifers of their initial As content over time [*Radloff et al.*, 2007; *Stute et al.*, 2007; *van Geen et al.*, 2008]. Such uncertainties make it difficult to speculate on the impact that the onset of irrigation pumping in recent decades may have had on the distribution of As in shallow groundwater. If the onset of irrigation pumping enhanced vertical recharge, the weight of the evidence would suggest that at least in Araihazar this resulted in lower rather than higher As concentrations in shallow aquifers [*Harvey et al.*, 2002, 2006; *Klump et al.*, 2006; *Stute et al.*, 2007].

6. Conclusion

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

[36] **Acknowledgments.** Columbia University and the University of Dhaka's research and mitigation work in Araihazar since 2000 has been supported by NIEHS Superfund Basic Research Program grant NIEHS 1 P42 ES10349, Fogarty International Center grant NIEHS 5-D43 TW05724-

01, NSF grant EAR 03-45688, NSF grant EAR 04-33886, and the Earth Institute at Columbia University. This is Lamont-Doherty Earth Observatory publication number 7170.

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