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Spatial variability of arsenic in 6000 tube wells in a 25 km^2 area of Bangladesh

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[1] Arsenic concentrations measured by graphite furnace atomic absorption range from < 5 to 900 μg/L in groundwater pumped from 6000 wells within a 25 km^2 area of Bangladesh. The proportion of wells that exceed the Bangladesh standard for drinking water of 50 μg/L arsenic increases with depth from 25% between 8 and 10 m to 75% between 15 and 30 m, then declines gradually to less than 10% at 90 m. Some villages within the study area do not have a single well that meets the standard, while others have wells that are nearly all acceptable. In contrast to the distribution of arsenic in the 8–30 m depth range which does not follow any obvious geological feature, the arsenic content of groundwater associated with relatively oxic Pleistocene sand deposits appears to be consistently low. The depth of drilling necessary to reach these low-As aquifers ranges from 30 to 120 m depth within the study area. *INDEX TERMS:* 1030 Geochemistry: Geochemical cycles (0330); 1045 Geochemistry: Low-temperature geochemistry; 1806 Hydrology: Chemistry of fresh water; 1831 Hydrology: Groundwater quality; 1884 Hydrology: Water supply; *KEYWORDS:* arsenic, groundwater, Bangladesh, tube well

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1. Introduction

[2] Untreated groundwater is the principal source of potable water for the significant fraction of the world's population that is not connected to a centralized water supply system, regardless of a country's wealth. In the United States, for instance, an estimated 42 million people (16% of the population) rely on their own private well for drinking water [*Solley et al.*, 1998]. In Bangladesh alone, about 130 million people (97% of the population) obtain their water from an estimated 10 million tube wells that have been installed across this predominantly rural country at an accelerating rate over the past 30 years [*Department of Public Health Engineering* (*DPHE*) *et al.*, 1999; *British Geological Survey and Department of Public Health Engineering*, 2001] (hereinafter referred to as BGS-DPHE). Although international organizations such as UNICEF in the 1970s and nongovernmental organization (NGOs) initially set the example by installing and promoting the use of tube wells, the vast majority of existing tube wells in Bangladesh were paid for privately by individual households [*van Geen et al.*, 2002]. A comparison of the cost of installing such a well (∼US$50 for a typical 50-ft well) with the per capita GNP of Bangladesh of US$1400, after adjusting for purchasing power [*World Bank*, 1999], is an indication of the popularity of groundwater as a source of pathogen-free drinking water. While the density of wells in Bangladesh may be particularly high, it probably accounts for only a fraction of the wells installed throughout South Asia, a region where accessible aquifers are widely distributed in sandy floodplain and delta deposits.

[3] Surveys of Bangladesh that followed the initial discovery of cases of arsenicosis attributable to elevated groundwater arsenic in the mid-1980s in West Bengal, India [*Chakraborty and Saha*, 1987], indicate that about one third of the existing wells yield groundwater that does not meet the local standard of 50 μg/L for arsenic in drinking water [*Dhar et al.*, 1997; *DPHE et al.*, 1999; BGS-DPHE; *McArthur et al.*, 2001]. Two thirds of the wells tested exceed the guideline value of 10 μg/L of the World Health Organization for arsenic in drinking water set in 1993 [*World Health Organization*, 1996]. There are troubling signs that other South Asian countries such as Vietnam, Cambodia, Pakistan, Myanmar, and Nepal are also significantly affected [*BGS-DPHE*, 2001; *Berg et al.*, 2001]. Even if the entire population of Bangladesh could be switched to low-arsenic water overnight, past exposure is

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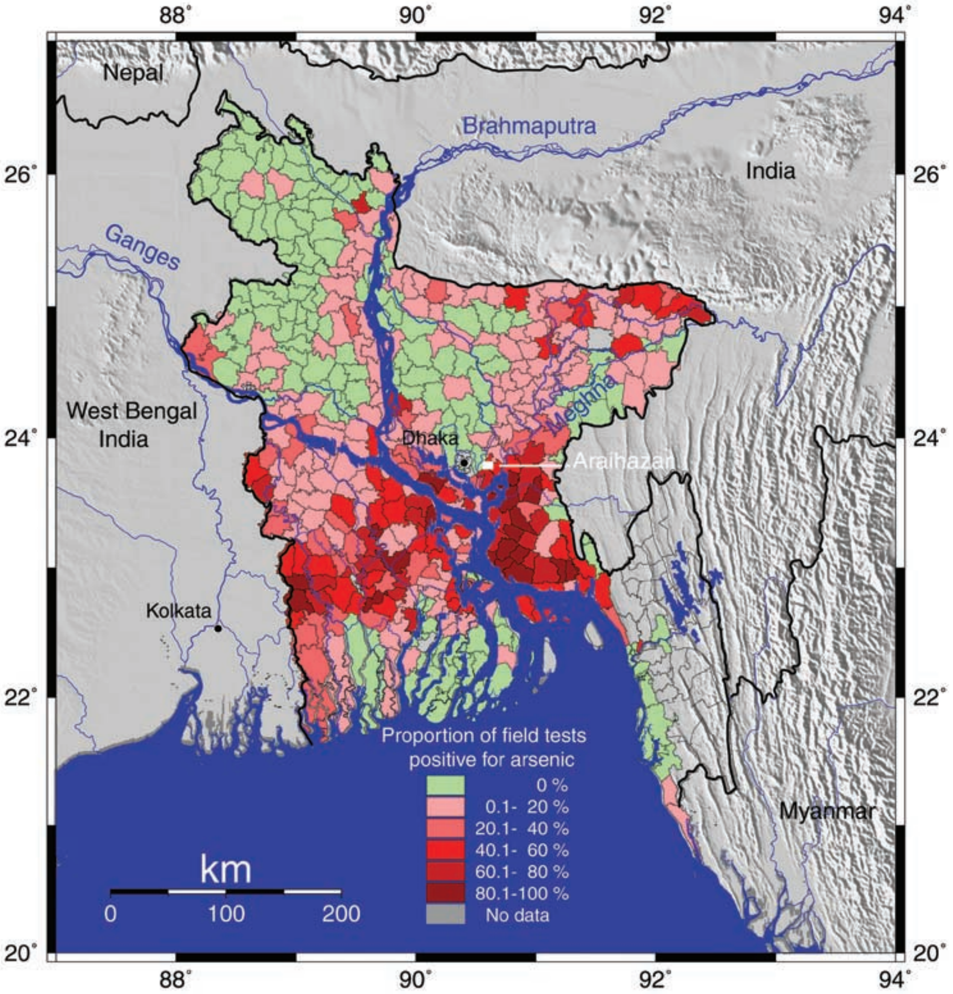


Figure 1. Proportion of tube wells that tested positive for As in upazillas of Bangladesh. Map prepared by J. W. Rozenboom (UNICEF-Dhaka) on the basis of 51,000 field tests (i.e., >100 μg/L) conducted across the country by DPHE/UNICEF in 1997–1999, substituted by with field tests for all wells in Sonargoan, Jhikargacha, Bera, Kachua, and Manikganj Sadar upazillas, and augmented with laboratory analyses (>50 μg/L) from the national survey of BGS-DPHE. Surrounding topography based on the GTOPO30 digital elevation model of the USGS EROS Data Center. Study area in Araihazar upazilla is indicated by a small white rectangle.

predicted to result in many thousands of additional deaths from various cancers each year, and this for the next several decades [*Smith et al.*, 2000; *Chen and Ahsan*, 2003; *Yu et al.*, 2003].

[4] A compilation of 51,000 field tests for arsenic conducted in Bangladesh by the Department for Public Health Engineering and UNICEF (Figure 1) shows that approximately one third of the country is essentially unaffected while over 60% of tube wells tested positive with a field kit (i.e., >100 μg/L [*Bangladesh Rural Advance Commission* (*BRAC*), 2000]). One of the vexing aspects of the mounting arsenic crisis in Bangladesh and elsewhere in South Asia is the bewildering degree of spatial variability of groundwater arsenic concentrations. Such sharp contrasts have convincingly been linked by various recent studies to the redox state of the underlying sediment with, broadly speaking, an association of low groundwater arsenic with relatively oxic, uplifted Pleistocene aquifers, and high groundwater arsenic concentrations in reducing Holocene aquifers [*DPHE et al.*, 1999; BGS-DPHE; *Nickson et al.*, 1998, 2000].

[5] In this paper, we document the spatial scale of arsenic variability for a transitional region in Araihazar upazilla, one of 460 similar administrative units in Bangladesh (Figure 1). DPHE/UNICEF field tests previously identified 75 high-As wells out of a total of 129 sampled within the 190 km^2 area of the upazilla. Araihazar is located 30 km east of Dhaka and marks the boundary between the uplifted Pleistocene Madhupur Terrace to the west that is unaffected by arsenic (no high-As tube wells out of 9 sampled by DPHE/UNICEF in neighboring Rupgang upazilla) and a large region to the south that is particularly affected (49 high-As wells out of 50 sampled in neighboring Homna upazilla). Additional sampling conducted in January 2000 guided us in the selection of a study area that contained tube wells spanning a wide range in As concentrations [*Zheng et al.*, 2003]. With 6000 tube wells covering an area of 25 km^2, the resolution of our geo-referenced data set is particularly

high. Every well within our study area was sampled and analyzed to set the stage for a coupled long-term study of the health effects of arsenic, as well as potential remedies (http://superfund.ciesin.columbia.edu). Although limited in comparison to the millions of wells distributed over the entire country, our data set is unique in the sense that ongoing nation-wide testing campaigns typically use a semi-quantitative field kit for arsenic. In addition, these campaigns generally do not take advantage of the Global Positioning System (GPS) to locate each well. Our sampling density in Araihazar is also considerably higher than in three special study areas (1 tube well per 7 km^2) examined as part of the highly informative country-wide work of *DPHE et al.* [1999] and BGS-DPHE. A few previous studies come closer to the level of detail of the Araihazar survey but they were limited to a single village [*Biswas et al.*, 1998; BGS-DPHE].

[6] The paper starts with a fairly detailed section on methods and quality control to demonstrate the integrity of this large data set of groundwater As concentrations. We then examine the spatial distribution of tube wells in relation to the subsurface geology of Araihazar and compare broad patterns in tube well arsenic to the previous nation-wide surveys. This is followed by a more detailed analysis of the arsenic distribution as a function of depth for the entire sampling area. We also examine more closely the As distribution in two 0.5 km^2 areas studied in detail in papers by Zheng et al. (manuscript in preparation, 2003) and Versteeg et al. (manuscript in preparation, 2003). These complementary studies allow us to extend the association of some key geological features with variations in groundwater As concentrations. The paper concludes with some suggestions for mitigation.

2. Methods

2.1. Sampling

[7] Water samples numbered 1–5000 were collected from 4999 contiguous tube wells in Araihazar upazila by six teams of Bangladeshi university graduates between March and June 2000. An additional set of 972 samples numbered 7001–8000 was collected by the same teams in November–December 2001. While one member of each team collected a water sample from a particular well, measured its position with a handheld GPS receiver, and identified it by affixing a numbered stainless steel plate, the other asked a household member a series of questions, including the date of installation, how the well was paid for, and the depth of the well. A household typically remembers the depth of its well because it had to pay for each 15-ft section of 1.5” ID PVC pipe that went into its construction. The accuracy of well positions determined before 1 May 2000 is estimated to be ±30 m, and ±10 m thereafter when the GPS signal was no longer degraded. Such a large and complex data set inevitably contains some gaps: GPS data are missing or obviously erroneous for 9 of the total of 5971 tube well samples that were collected; depth information is missing for 136 samples.

[8] After continuous hand-pumping at a rate of 20–35 L/min for at least five minutes, i.e., for long enough to flush several times the 16 L internal volume of a typical 15-m (50 ft) well, a polyethylene graduated cylinder cut off to contain 60 ± 2 mL when overflowing was rinsed several times with well water and then used to fill a high-density polyethylene Nalgene bottle that had been acid-cleaned and rinsed several times with Milli-Q water. The samples were not filtered based on our experience that in most cases the screen installed at the bottom of each well effectively excludes particles of aquifer material [*Zheng et al.*, 2003]. To the set of 20 sample bottles taken to the field by each team every day, 1 mL of 7 N Optima HCl was added in advance in our local laboratory for sample preservation. An extra three bottles prepared with the same acid were added to each daily set of 20 samples for quality control, one of which was in addition spiked with 0.6 mL of a 50 mg/L As standard solution to determine recovery. The purpose of these additions was to identify potential artifacts in the data set caused by poor sample preservation or analysis. One of the extra bottles taken to the field for quality control was filled with 60 mL of distilled water from Dhaka University as a blank.

[9] In the field, sets of adhesive labels with preprinted ID numbers were attached to each sample bottle and questionnaire at the time of collection. To reduce the chance of transcription errors from the questionnaire, well ID, GPS position, and well depth were also marked directly on the bottle. Three sample bottles were filled with water from the first tube well sampled by each of the six teams on any given day: the first two of these bottles were replicate samples; the third bottle contained the As spike. A total of 353 sets of replicates, consistency standards, and blanks distributed fairly evenly across the 5971 well samples were collected by the six teams.

2.2. Analytical Methods

[10] All samples were analyzed for As by graphite furnace atomic absorption (GFAA). The majority of well samples (4,821) and associated quality control samples were analyzed at Lamont-Doherty Earth Observatory (LDEO) with a Hitachi Z8200 instrument by diluting each sample 1:5 in a matrix modifier solution containing 50 mg/L Ni and 2% by volume Optima HNO_3. Instrument response was linear up to an absorbance of 0.3, corresponding to a concentration of 150 μg/L As (750 μg/L in the undiluted sample). A total of 35 well samples containing over 750 μg/L (most of these were samples spiked with As) were analyzed again at a 1:10 dilution. Instrument drift in sensitivity was corrected by linear interpolation between sets of standards prepared in pure (i.e., without sample) matrix modifier solution included at the beginning and at the end of each set of ∼50 samples. We conservatively estimate a detection limit of 1 μg/L by this method (5 μg/L in the undiluted sample) taking into account day-to-day variations in the reproducibility of a blank determination for a pure Ni/HNO_3 matrix modifier solution as a function of graphite tube and lamp condition. The arsenic concentration of blanks of distilled water from Bangladesh was always below the detection limit, with the exception of one bottle that had been spiked and another that contained a replicate groundwater sample rather than distilled water.

[11] The remaining 1,150 well and quality control samples were analyzed at the Mailman School of Public Health (MSPH) on a Perkin Elmer AAnalyst 600 instrument by 1:5 dilution in a 0.2% HNO_3 matrix modifier solution with

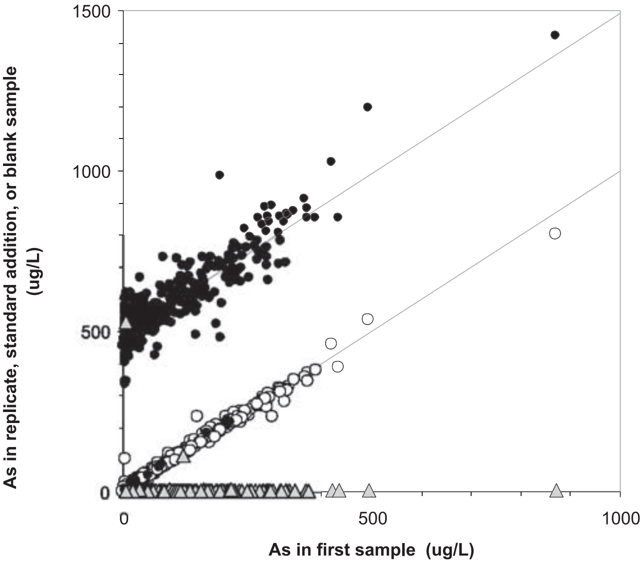


Figure 2. Compilation of arsenic analyses for all quality control samples. Arsenic measurements for well samples collected as replicates (open circles) and for standard additions in the field (solid circles) and blanks (shaded triangles) are shown as function of the As concentration measured in the first sample from each set of quality control samples. The expected trends are indicated by two solid lines with a slope of 1, the first passing through the origin for replicates and the other with an intercept of 492 μg/L As corresponding to the standard addition. There are a limited number of obvious discrepancies relative to these trends. The concentration of one replicate sample increased from <5 μg/L to 200 μg/L over the course of a year, while the arsenic content of the first sample from the same well was below the detection limit. This suggests the replicate contained a particle of sediment that gradually released arsenic after it was acidified. In addition, an As spike was mistakenly added to a blank instead of a replicate sample; one blank bottle was filled with a replicate sample; and arsenic spikes were not added to 12 out of the 265 sets of quality control samples.

Pd and $Mg(NO_3)_2$ added as matrix modifiers. No sample from this set exceeded the linear range of the instrument (250 μg/L at 0.3 absorbance). Analysis of Standard Reference Material 1643d (Trace elements in Water) from the National Institute of Standards and Technology certified to contain 56 μg/L As included with these runs averaged a value of 61 ± 1 μg/L (n = 6). Intercalibration between the two laboratories with a subset of 24 samples covering the <5 to 800 μg/L range showed no systematic difference between the two procedures. The intercalibration showed agreement within 50 μg/L for all but two samples, for which differences of 66 and 80 μg/L corresponded to 12 and 14% of the total As concentration, respectively. The two data sets were therefore combined.

2.3. Data Quality Control

[12] Sets of replicate and standard addition samples, as well as blanks, were typically included in the same run; occasional outliers were analyzed a second time. Overall, the quality control data show the expected trends: replicate samples generally follow the expected 1 to 1 relation; the As concentration of samples with standard additions is offset from this relation by the expected amount (492 μg/L), although with significantly more scatter (Figure 2). Excluding one outlier apparently due to a sampling artifact, the standard deviation for the difference between replicates is 10 μg/L (n = 353), with no apparent dependence on the absolute As concentration (Figure 3). This standard deviation reflects the error from two measurements; we therefore estimate the standard deviation for an individual measurements to be $\sigma_{meas} = 10/\sqrt{2} = 7$ μg/L. Reproducibility was somewhat better for the method/instrument combination used at MSPH (±4 μg/L) than at LDEO (±11 μg/L). The good match for all but one of the sets of replicates is consistent with the presence of As in tube well water in the dissolved (or perhaps colloidal) state. This confirms that the slotted screen and the gravel pack installed at the bottom of each well effectively remove aquifer particles whose As contribution to a sample upon acidification would have been highly variable.

[13] To calculate the increase in arsenic concentration from standard additions in the field, the concentration of the spiked sample was first multiplied by a factor of 61 mL/61.6 mL to account for dilution by the As spike itself. From this value, the mean of the two replicates from each well was then subtracted. Excluding the 12 sets of quality control samples where the As spike was clearly not added, this difference averages 498 μg/L, with a standard deviation of 56 μg/L (n = 341), i.e., a relative standard deviation of 0.112, or 11% (Figure 2). The difference is just within two

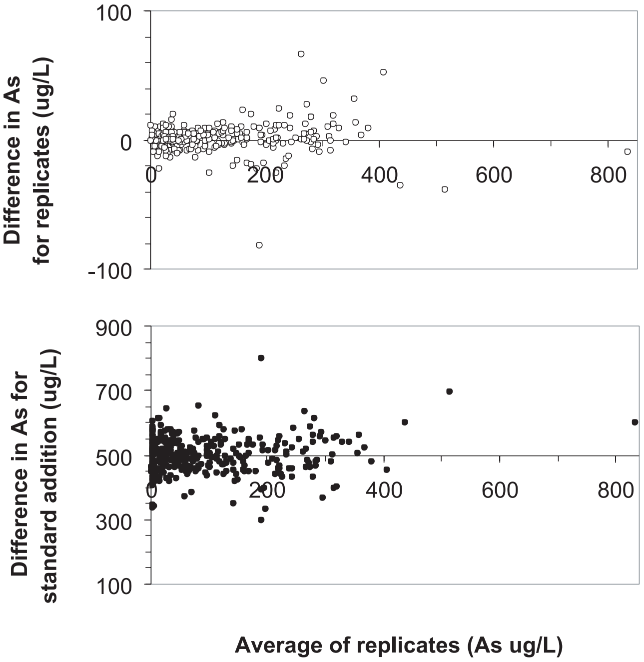


Figure 3. Variability of replicate determinations and standard additions. (top) Differences in As concentration between replicate measurements as a function of the mean of the replicates. (bottom) Difference in concentrations between the spiked sample and the mean of the two replicate determinations for the unspiked sample, again as a function of the mean of the replicates. Clear outliers identified in Figure 2 are excluded in Figure 3.

standard deviations of the error of the mean (56 μg/L/$\sqrt{341}$ = 3 μg/L) from the expected value of 492 μg/L calculated from the concentration of the spike. This indicates that, on average, the analytical procedures that were followed eliminated any systematic bias in instrument response for standards prepared in pure matrix modifier relative to groundwater samples diluted in matrix modifier. For these measurements too, reproducibility of the difference in As concentration between spiked and unspiked samples was somewhat better at MSPH (±37 μg/L) than at LDEO (±62 μg/L for quality control analyses associated with samples 1–5000 and ±43 μg/L for samples 7001–8000, excluding one outlier).

[14] The variability of the increase in concentration due to standard addition reflects several sources of error: imperfect control of sample and/or spike volume in the field, day-to-day differences in instrument calibration, as well as potential matrix effects that could be related to the composition of a specific groundwater sample (e.g., ionic strength, other groundwater constituents). To test for matrix effects, a subset of 8 samples for which the effect of As additions in the field deviated significantly from the expected value were reanalyzed at LDEO, with individual calibration for each sample by the method of standard additions to the sample cup. The slopes of these calibrations, constrained by As addition of 150, 300, and 500 μg/L, varied around the mean with a standard deviation of 7% while the uncertainty of the individual slopes ranged between 0.5 and 2.5%. This suggests that a significant portion of the 11% relative standard deviation for As recovery from additions in the field is probably due to a relatively small but detectable matrix dependence of the response of the instruments.

[15] A simple model described in Appendix A quantifies the impact of this effect on the uncertainty of the well As data, obtained in the majority of cases with a single measurement and without standard additions in the field or in the laboratory. According to this model, an absolute uncertainty in measured As concentration of 7 μg/L dominates below 70 μg/L. Above this concentration, an additional relative error of ~10% due to differences in sample matrix becomes the larger source of uncertainty. Although greater accuracy could have been achieved with multiple-point standard additions performed in the laboratory for each sample, we felt the marginal improvement would not have justified the additional effort required, considering the two orders of magnitude dynamic range in measured As concentrations.

3. Results

[16] An IKONOS satellite image of our study area in Araihazar was purchased from Space Imaging Inc. to map the tube well As data. This image, taken in November 2000, provides reflectance data in four wave bands at 4-m spatial resolution and panchromatic data at 1-m resolution (http://www.spaceimaging.com). The data were processed to emphasize in purple the presence of water in streams, ponds, and irrigated fields without vegetation (Figure 4). Contrasting bright areas of the IKONOS image indicate dry, presumably uncultivated fields. Green areas show the vegetation of the fields that are cultivated as well the tree cover that identifies the villages. Overlay of the GPS data on the satellite image indicates that most tube wells are clustered in villages. The largest stream meandering in a southerly direction through our study area is the so-called Old Brahmaputra River. The name underlines the fact that the much larger Brahmaputra River used to flow through the region until a major shift in its course in the 1800s [*Ferguson*, 1863], possibly triggered by a major earthquake. A 10-km long series of villages follows the inner bank of the main meander of the Old Brahmaputra River; the shape of some other series of villages further inland suggest their possible association with the location of natural levees in the past. Villages and fields share the available land in roughly equal proportion in our study area, where the population density of 2600/km^2 is nearly three times higher than the national average. Significantly, responses to a questionnaire collected with the well samples indicate that the vast majority of these wells were paid for by individual households and therefore are privately owned [*van Geen et al.*, 2002]. The survey data also indicate that half the existing wells in the study area were installed between 1995 and 2000, i.e., after the discovery of the epidemic of arsenicosis in Bangladesh.

[17] The wide range of As concentrations spanned by tube wells sampled within Araihazar (<5–860 μg/L) is similar to that reported for laboratory analyses of water from 3534 tube wells distributed across the entire country [*DPHE et al.*, 1999; *BGS-DPHE*, 2001]. Of the nearly 6000 samples of tube well water from Araihazar that were analyzed, 25% contain ≤10 μg/L As, the WHO guideline value for As in drinking water (Figure 5). Most of the wells with the lowest As concentrations are located in the northwestern portion of the study area (Figure 4). The As concentration of another 22% of the samples with higher concentrations still meets the standard for drinking water in Bangladesh of 50 μg/L, which was until recently also the standard set by the U.S. Environmental Protection Agency. Wells in this secondary category are more widely distributed throughout the study area. The number of wells with As concentrations in the 51–100, 101–200, and 201–900 μg/L ranges is comparable and accounts for 18, 19, and 15% of the data set, respectively (Figure 5). Unlike the central region of the study area where arsenic concentrations are highly variable, there are very few wells supplying water that meet the Bangladesh standard for As to inhabitants of the western and easternmost areas (Figure 4).

[18] The overall depth distribution of As within Araihazar is also remarkably similar to that reported by BGS-DPHE for the entire country. In this paper, depths are listed in both meters and feet because the data were collected, and are locally referred to, in feet. Although there is considerable scatter in As over the entire 8 to 90 m (25–300 ft) depth range of wells in Araihazar, concentrations greater than 400 μg/L are largely restricted to the 12–24 m (40–80 ft) depth range (Figure 6). Unfortunately, this is also the depth range for two thirds of all tube wells in the study area. While the correspondence is disturbing, it does not necessarily indicate a causal relation between groundwater As levels and the density of wells. The average As concentration increases steadily from 40 μg/L in the 8–12 m (24–36) ft interval to 160 μg/L in the 20–30 m (65–100 ft) interval (Figure 6). Across the same depth intervals, the proportion of wells that yield water containing over 50 μg/L As rises from 25 to 80%. Between 30 and 90 m (100 and

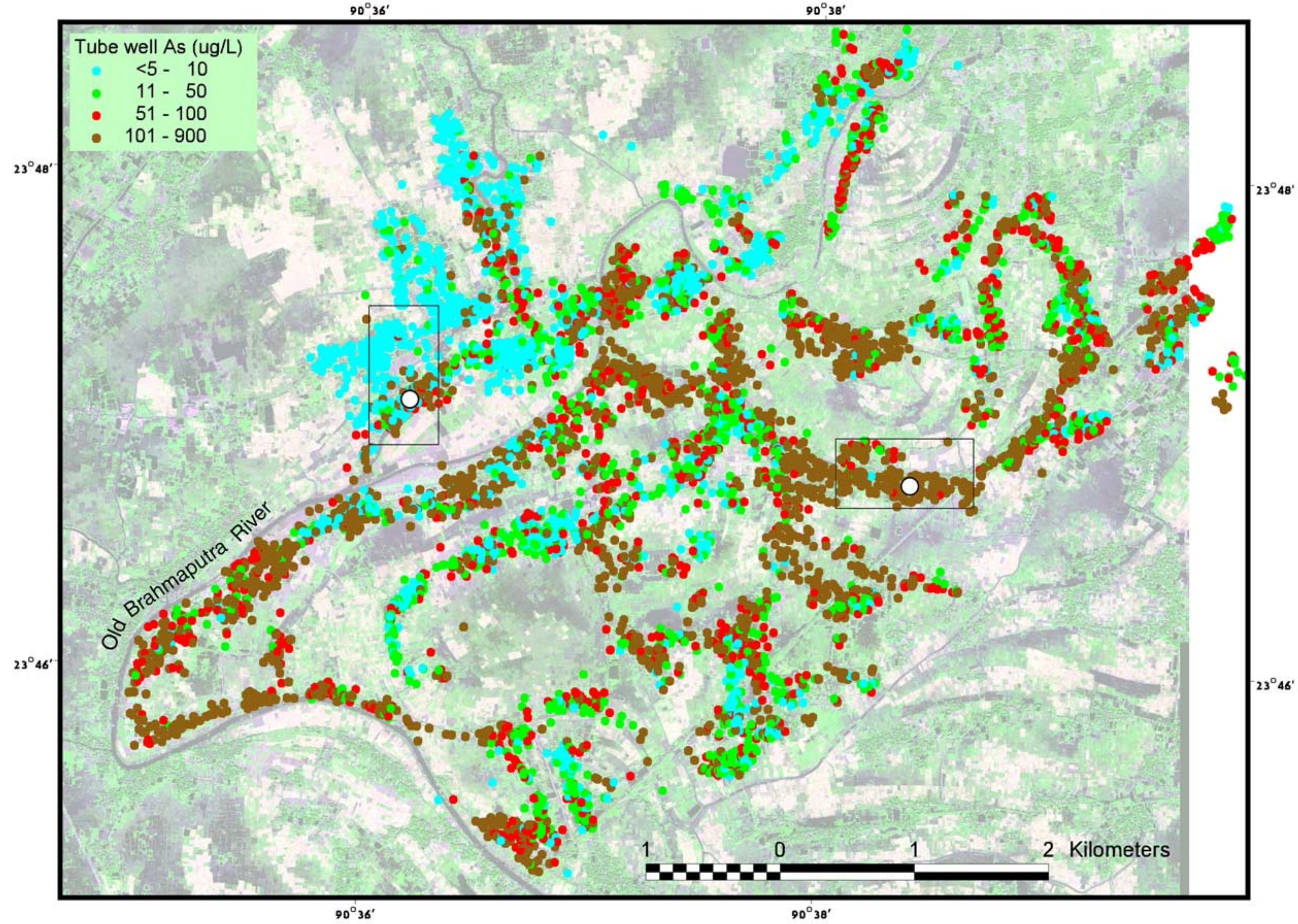


Figure 4. Distribution of As in 6000 tube wells of Araihazar. The IKONOS Space Imaging Inc. satellite image emphasizes the presence of water at the surface in purple and dry land in white. The two rectangular areas are shown as close-ups in Figures 9 and 10 are discussed in more detail by Zheng et al. (manuscript in preparation, 2003) and Versteeg et al. (manuscript in preparation, 2003). The locations of the two sediment cores collected within these two areas are indicated by a white circle.

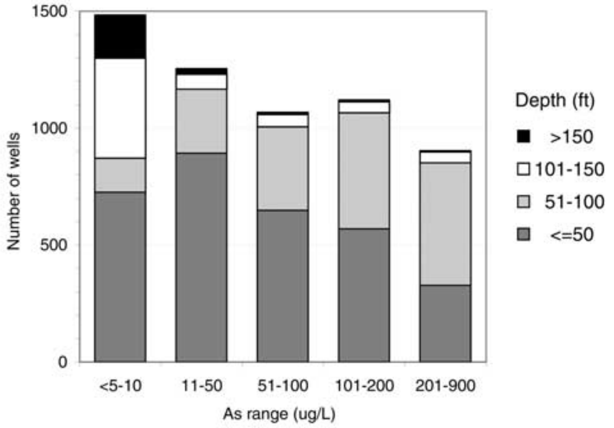


Figure 5. Histogram of number of tube wells in different As concentration and depth ranges.

300 ft) depth, which includes 15% of all wells in the study area, the proportion of wells that do not meet the Bangladesh standard for As in drinking water declines steadily from 29 to 8%. Although As concentrations in groundwater from the majority of wells shallower than 30 m (100 ft) exceed 50 μg/L, and from the majority of wells deeper than 30 m are below 50 μg/L, a significant fraction of the wells does not fit this simple pattern.

[19] The As distribution in Araihazar groundwater becomes a little less bewildering if depth, grouped in four intervals, and location are considered simultaneously (Figure 7). The shallowest of these surfaces corresponds to depths up to 15 m (50 ft) and shows several clusters of villages where As concentrations are below 50 μg/L with relatively few exceptions. These clusters appear to be grouped in two approximately linear arrays on either side of the northeast to southwest-trending course of the Old Brahmaputra river through the northern half of the study area (Figure 7a). There appears to be little extension of these features to the next deeper surface, between 15 and 30 m (50 and 100 ft) depth, where water yielded by the vast majority of wells contains over 50 μg/L As (Figure 7b). The number of wells is much lower in the next depth interval, between 30–45 m (100–150 ft), with the exception of the northwest portion of the study area where almost wells extend deeper than 30 m (100 ft). Another distinguishing feature of the northwestern region of the study area is that most wells contain less than 10 μg/L As, while As concentrations in most wells elsewhere within the 30–45 depth range are higher than 50 μg/L, and often higher than 100 μg/L (Figure 7c). Below 45 m (150 ft), the density of well remains highest in the northwestern portion of the region, but a significant proportion of the wells in the remaining area also meet the local drinking water standard of 50 μg/L (Figure 7d).

4. Discussion

4.1. Temporal Changes

[20] Although the limited number of well-documented time series that are available do not indicate any large

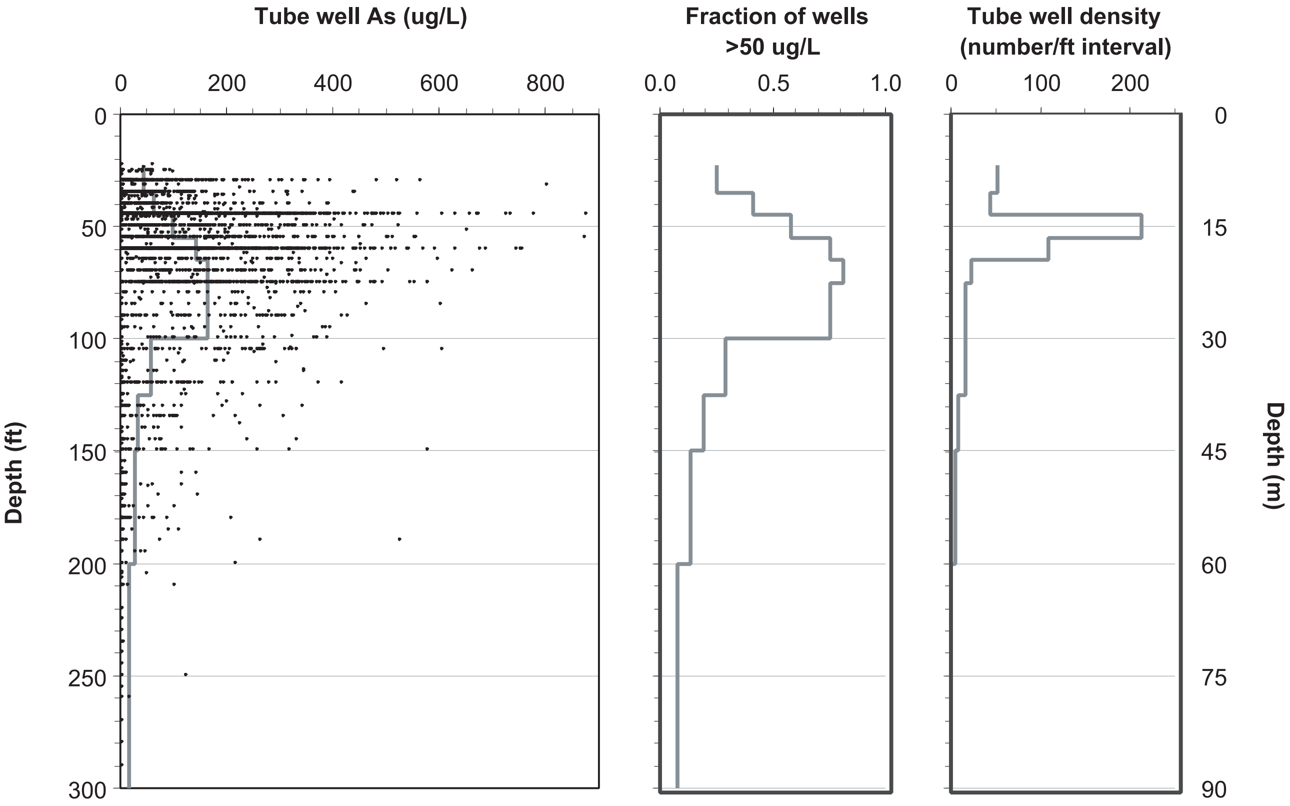
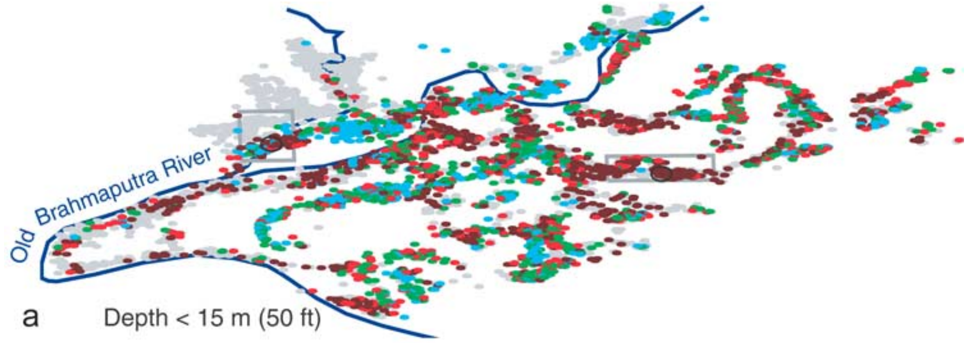
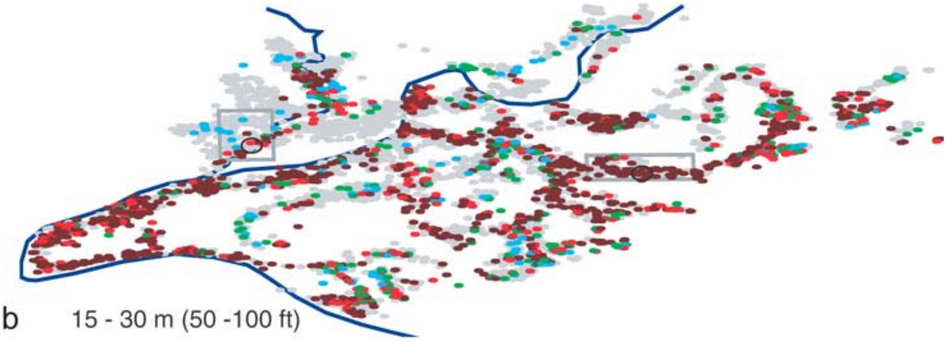


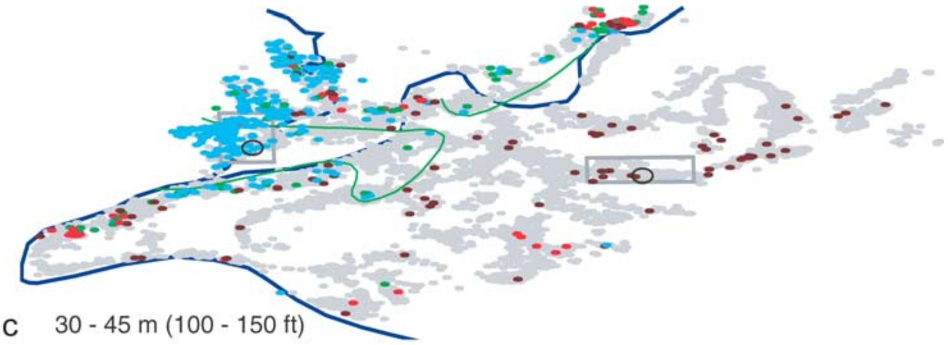
Figure 6. Depth distribution of As in nearly 6000 tube wells of Araihazar. (left) Solid lines indicate average As concentrations for various depth intervals. (middle) Proportion of wells containing over 50 μg/L As. (right) Density of wells per ft of depth range for the same intervals.



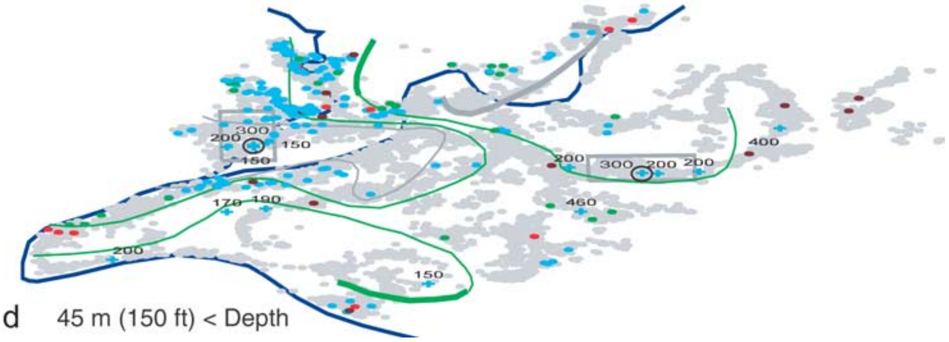
a Depth < 15 m (50 ft)



b 15 - 30 m (50 -100 ft)



c 30 - 45 m (100 - 150 ft)



d 45 m (150 ft) < Depth

Figure 7. Distribution of tube well As in Araihazar for four depth intervals. The color scale of the dots is the same as in Figure 4. Grey dots indicate the position of all wells. Distorted boxes correspond to the close-up areas identified in Figure 4 that are enlarged in Figures 9 and 10; black circles indicate the location of sediment cores discussed by Zheng et al. (manuscript in preparation, 2003). The location of community wells is indicated by crosses, together with their corresponding depths (in feet). Green contours mark areas where wells consistently meet the Bangladesh standard at depths of 30 m (100 ft) and depths >45 m (150 ft), respectively.

temporal trends for groundwater As in Bangladesh [BGS-DPHE; *van Geen et al.*, 2002], there is a worrisome trend in our results from Araihazar that suggests that As concentrations may slowly increase as a tube well ages. Regressions of As concentration as a function of year of installation indicate small but statistically significant decreases in the mean concentration in several depth intervals (Figure 8). This corresponds to an equivalent increase in As concentration with well age. Since the individual slopes were statistically not different from each other, a

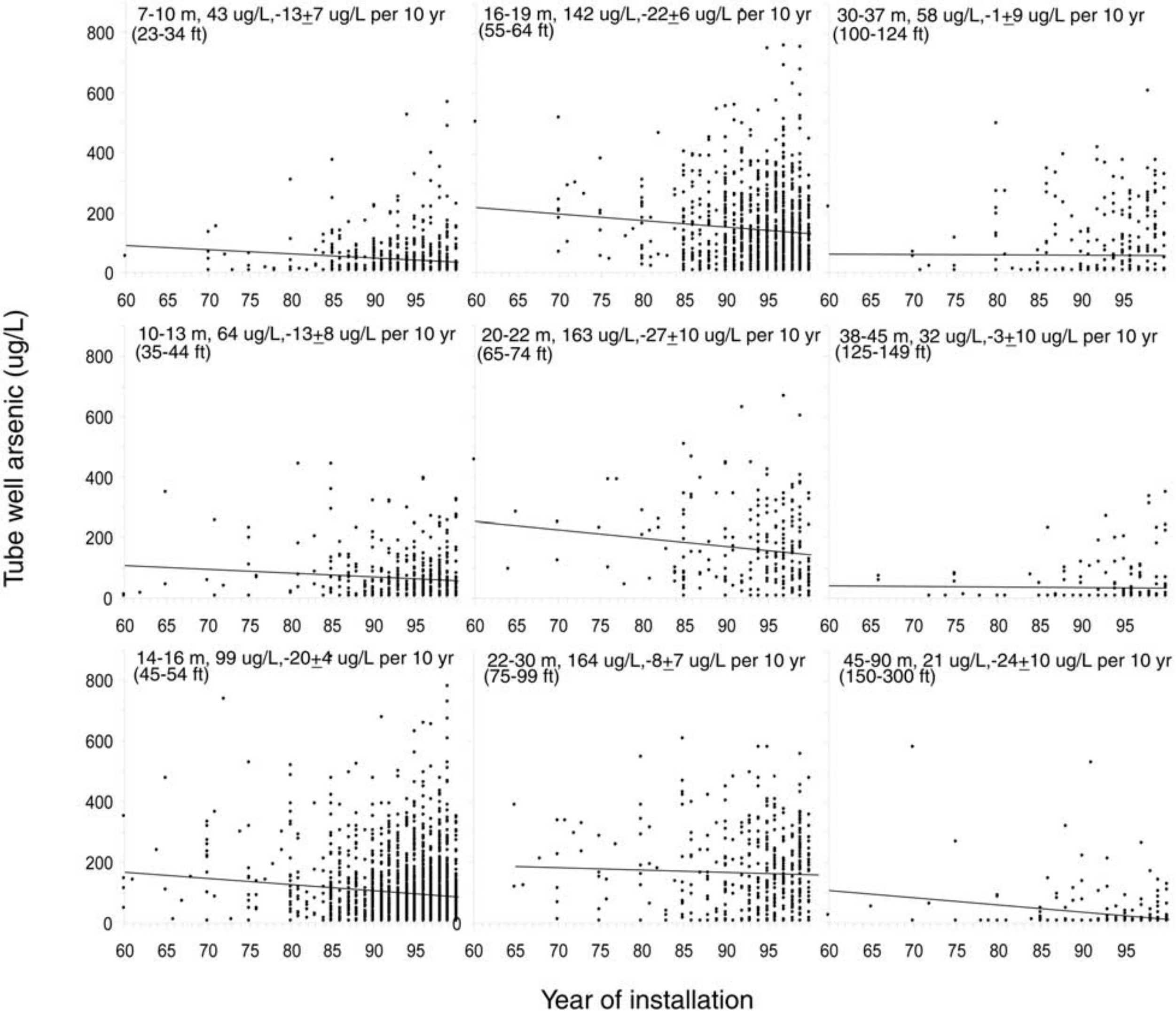


Figure 8. Regressions of As concentration as a function of year of installation in nine different depth intervals. The same intervals shown in Figure 6 were used, with the exception of the two deepest intervals which were combined into one (150–300 ft) in this analysis. The depth range, the mean arsenic concentration within that range, and the slope of the regression are listed for each panel.

pooled estimate of an increase of 16 ± 2 μg/L in well As per decade of well age was calculated by imposing a common slope on the different depth intervals, while allowing for different means. Essentially the same result is obtained for regressions on the log of the As concentrations. The pooled slope estimate of the slope indicates an increase of 0.019 ± 0.003 in log As concentration per year. This suggests that, on average, As levels may have increased by about 2% per year when comparing wells from the same depth interval within our study area. Such a small increase is not inconsistent with several time series of groundwater As from Araihazar that do not show a detectable change because these span only a single year at this point [*van Geen et al.*, 2002]. *Burgess et al.* [2000, 2002] also noted increases in groundwater As as a function of well age on the basis of the BGS-DPHE data, but without taking depth into account or the possibility that the mean depth of installation of wells has changed over time. The explanation proposed by *Burgess et al.* [2000] is that, given a patchy As distribution at depth, the likelihood of drawing high As water from a particular well increases as the integrated amount of water pumped from a given well increases. Potential changes in groundwater As concentration over longer timescales therefore cannot be excluded at this point. Invoking a different scenario linking As mobilization to the penetration of labile dissolved organic matter in shallow aquifers, *Harvey et al.* [2002] recently suggested that groundwater As concentrations may have increased as a result of increased water withdrawal for irrigation. The ensuing controversy regarding this important issue [*van Geen et al.*, 2003b] underlines the need for establishing As baseline levels in potentially sensitive areas and continued monitoring of such sites.

4.2. Groundwater As and Local Stratigraphy

[21] A closer examination of two contrasting 0.5 km^2 regions within the study area illustrates the relation between the main features of the As distribution and local geology. The first of these areas covers the transition from the northwest, where wells are almost all deeper than 30 m (100 ft) and low in As, to a more complex regime to the

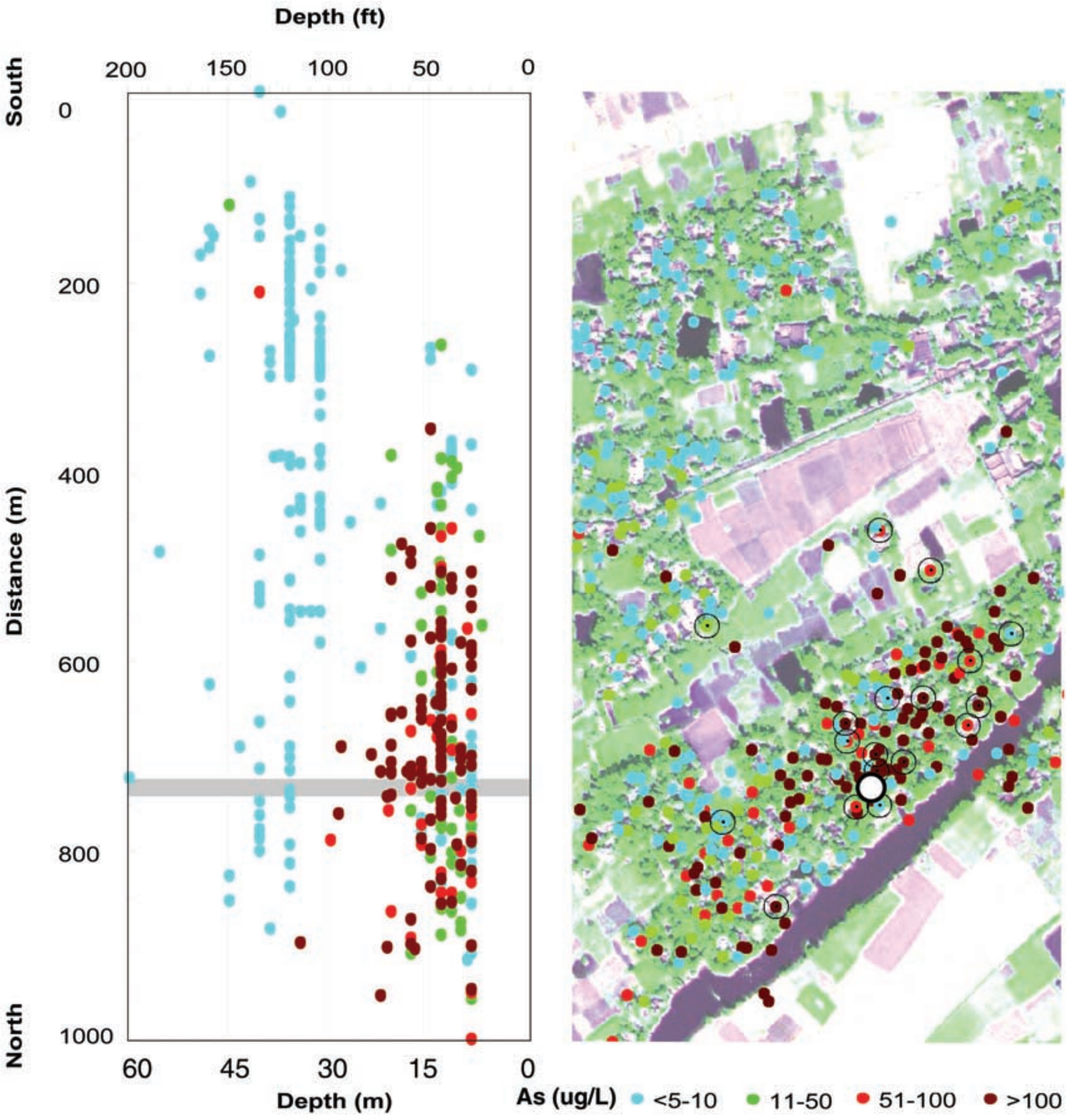


Figure 9. (right) Geographic distribution of As and (left) depth section of As in first close-up area in Figure 4. Black circles indicated samples with more detailed information discussed by Zheng et al. (manuscript in preparation, 2003). White dot (right) and gray bar (left) indicate the location of the sediment core.

south (Figure 9). In this southern portion of the close-up area, wells extending below 30 m (100 ft) depth are low in As in the vast majority of cases. Tube wells from the same region at shallower depths, however, are highly variable in As and do not exhibit any clear spatial pattern. There is no such contrast between As concentration in shallow and deep wells in the second close-up area where elevated As concentrations extend to 60 m (200 ft) depth (Figure 10). Most wells contain over 100 μg/L As in this village and only a handful meet the local standard for As drinking water of 50 μg/L.

[22] The north–south section of groundwater As in the first of the close-up areas shows that there are few wells with reported depths between 21 and 30 m (70 and 100 ft) throughout this region (Figure 9). Zheng et al. (manuscript in preparation, 2003) describe a sediment core collected inside the village near the small stream that flows through the area. This core contains a thick impermeable clay layer between 15 and 30 m (50–100 ft), with sandy material above and below. Resistivity measurements mapping the subsurface in the same area, described by Versteeg et al. (manuscript in preparation, 2003), are consistent with the presence of a thick clay layer that extends largely uninterrupted throughout the village and the surrounding fields. Additional resistivity sections (Versteeg et al., manuscript in preparation, 2003) indicate that the same clay layer gradually shoals toward the north and outcrops in the region where there are no shallow wells. Combining this information with the well data indicates that an impermeable and tilted layer separates shallow aquifers with variable As levels from a deep aquifer containing in most cases <10 μg/L As in this particular region. An attempt to radiocarbon date organic matter extracted from a peat layer within the clay layer at the site

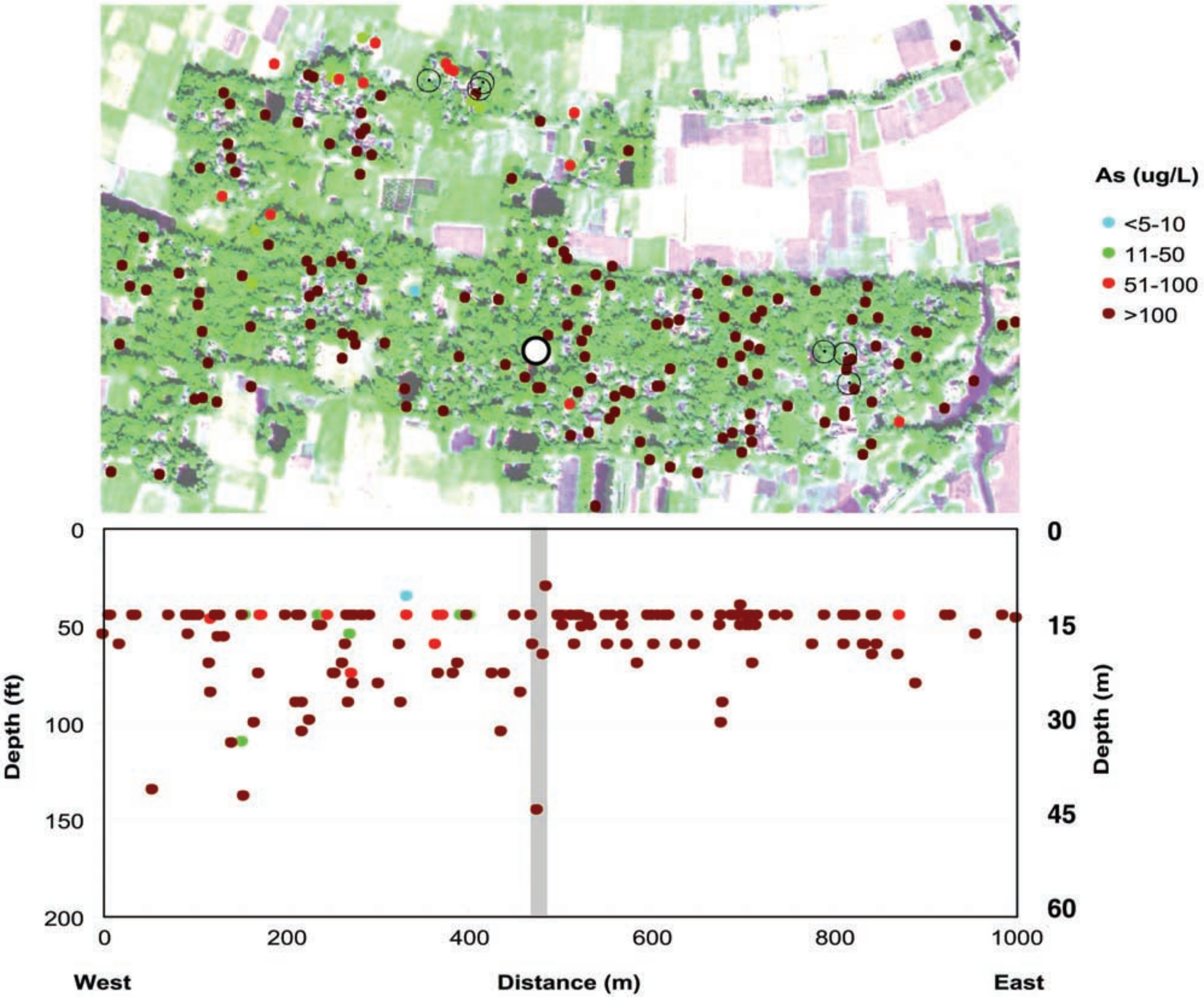


Figure 10. (top) Distribution of As and (bottom) depth section of As in second close-up area shown in Figure 4. Black circles indicate samples with more detailed information discussed by Zheng et al. (manuscript in preparation, 2003). Location of sediment core collected in this area indicated by a white circle (top) and a gray bar (bottom).

of the core yielded an age greater than 40,000 years (Zheng et al., manuscript in preparation, 2003).

[23] A sediment core was also recovered from the second close-up area with mostly high-As wells (Figure 10). Although it is located only 3 km east of the first core, no clay layer of comparable thickness was recovered. Instead, three 5 m (15 ft) thick clay layers evenly spaced between 30 and 60 m depth were observed (Zheng et al., manuscript in preparation, 2003). A pair of overlapping resistivity sections collected from the same area suggests that, in addition to being thinner, these layers do not extend north or south beyond the village (Versteeg et al., manuscript in preparation, 2003). In contrast to the first close-up area, radiocarbon dating of organic carbon in the village with mostly high-As wells indicates that the entire sediment section recovered from this location was deposited after the last glaciation ended about 20,000 years ago and sea level started to rise (Zheng et al., manuscript in preparation, 2003). Low groundwater As concentrations in water from a 90-m (300 ft) deep community well at this site [*van Geen et al.*, 2003a] indicates that deep aquifers do not have to be over 40,000 years old to yield safe water, at least in Araihazar. The wide range of As concentrations in shallow aquifers of the first close-up area and low-As levels in the deepest sampled aquifer in the second area suggest factors other than the age of a sandy deposit, possibly related to the nature of the solid phase, play a role in controlling groundwater As concentrations.

4.3. Groundwater As and Characteristics of the Solid Phase

[24] Iron oxyhydroxides that coat aquifer particles appear to be the solid phase that, in most cases, controls the As concentration in Bangladesh groundwater. Because As has a very strong affinity for Fe oxyhydroxides, the presence of such coatings under oxic conditions keeps groundwater arsenic levels low [*Bhattacharya et al.*, 2001; *Nickson et al.*, 1998, 2000; *Cummings et al.*, 1999; *McArthur et al.*,

2001; *Harvey et al.*, 2002; *Zheng et al.*, 2003]. Under the reducing conditions that are often attained in river floodplain and delta sediments, however, microbially-mediated dissolution of such oxyhydroxides can result in large increases in the concentration of dissolved As. In this context, it is worth noting that enriching groundwater by e.g., 200 μg/L in As corresponds to a loss of only ~0.1 μg/g from the solid phase, a loss that would be hard to detect relative to the average As concentration in the continental crust of 2 μg/g. While the As content of sediment supplied by the Ganges, Brahmaputra, and Meghna rivers is not particularly high, 1–6 μg/g according to *Datta and Subramanian* [1998], a small labile fraction associated with Fe oxyhydroxides would clearly be sufficient to considerably elevate groundwater As concentrations. Indeed, unexceptional As concentrations in the solid phase were observed in the BGS-DPHE special study areas and our two cores from Araihazar (Zheng et al., manuscript in preparation, 2003). The underlying assumption of this discussion is therefore that As is present in aquifer material throughout the study area in sufficient amounts to potentially cause a large increase in groundwater As upon transfer to the dissolved phase. Rather than conducting an elusive search for As-enriched sediment to explain the distribution of groundwater As in Araihazar, we therefore interpret the As distribution as primarily a reflection of the variability of redox conditions in the host aquifer.

[25] How do redox conditions vary in the sediment cores collected in the two close-up areas? A useful albeit qualitative indicator of the redox state of a sandy aquifer is color. This has been noted on the basis of boreholes distributed throughout of Bangladesh where shallow gray sediment deposited since the last glaciation contrast with deeper Pleistocene sands that are bright orange/brown in color (BGS-DPHE). Within the first of the close-up areas in Araihazar, for instance, undetectable radiocarbon in the dividing clay layer and the orange color of the sands underneath are both consistent with an association of the low-As groundwater with Pleistocene sands, most likely the Dupi Tila Sand formation of Pleistocene age (BGS-DPHE; Zheng et al., manuscript in preparation, 2003). The stratigraphy of the first close-up area is not unlike that of the special study area in Lakshmipur located further south and described by BGS-DPHE, including the presence in that area of a peat layer without detectable radiocarbon. Taking into consideration the geologic context of the study area, we infer that the thick clay layer underlies floodplain and deltaic sediment deposited across the Ganges-Brahmaputra-Meghna delta since sea level returned to its present state during the Holocene, about 10,000 years ago [*Umitsu*, 1993; *Goodbred and Kuehl*, 1999, 2000; *Goodbred et al.*, 2003].

[26] Although the origin of the color contrast between Pleistocene and more recently deposited sediment has not been fully investigated, it is probably related to the proportion of pure Fe(III) oxyhdroxides and mixed Fe(III)/Fe(II) oxyhydroxides and other Fe(II) phases. Mixed Fe(II)/(III) oxyhydroxides such as magnetite [*Lovley*, 1990], for instance, are typically black while the color of pure Fe(III) oxyhydroxides such as goethite or hematite ranges from yellow to red [*Cornell and Schwertmann*, 1996]. Other Fe(II) phases such as siderite or vivianite may also be present, but these are typically not black. Overall, the association of high groundwater As with gray sediment may therefore reflect a relation between sediment color, i.e., the redox state of Fe in the particulate phase, and the extent of dissolution of Fe hydroxides naturally enriched in As. This would be consistent with groundwater data within Araihazar [*Zheng et al.*, 2003, manuscript in preparation, 2003] and throughout Bangladesh (BGS-DPHE) showing a general, yet scattered, relation between elevated dissolved As and Fe concentrations in groundwater. It is worth noting in this context that even the groundwater pumped from the Dupi Tila Sand formation is anoxic, at least in our study area (Zheng et al., manuscript in preparation, 2003). Reference to this aquifer as an oxic Pleistocene deposit should therefore be related to the oxidation state of Fe on the solid phase rather than the oxygen content of groundwater.

[27] Further analysis of the core from our second close-up area indicates that the relation between groundwater As and sediment color may be more complicated. Sediment from the entire 90 m (300 ft) section at this location ranges between different shades of gray. This is consistent with the association of gray color with deposition over the past ~10,000 years of the Holocene documented elsewhere in Bangladesh (BGS-DPHE). But As in concentrations in groundwater from a set of monitoring wells installed around this core range widely from 10- to 500 μg/L. Although orange or brown sediment are rather consistently associated with groundwater low in As, a gray color therefore does not necessarily indicate elevated dissolved As concentrations.

4.4. Groundwater As and Regional Geology

[28] The origin of the sharp contrast in both stratigraphy and groundwater As levels between the two close-up areas where sediment cores were collected is not entirely clear at this point. A statistical analysis of the entire data set of groundwater As into a series spatial clusters of about 60 wells each shows that the contrast is representative of a regional boundary that runs diagonally through the study area from the southwest to the northeast (A. Gelman et al., submitted manuscript, 2002). Northwest of this division, a “safe depth” below which groundwater As levels are typically low can be calculated for most of the clusters of wells, together with a measure of the uncertainty of this estimate. Southeast of the same division, existing wells do not indicate a deep aquifer that is consistently low in As in most clusters. The explanation for the contrast between the two regions could be that relatively oxic Pleistocene aquifers were uplifted in the northwest portion of the study area together with the Madhupur Terrace that extends over a considerable area toward Dhaka [*Goodbred and Kuehl*, 2000; BGS-DPHE]. Another possibility, which does not exclude the first, is that the thicker Holocene layer in the eastern portion of the study area reflects infill of a major river channel formed during the previous glacial low-stand in sea level about 22,000 years ago. Similar pronounced gradients in the depth to relatively oxic and low-As aquifers of the Dupi Tila formation over a distance of a few kilometers have also been documented in the Chapai Nawabgnaj special study area located northwest of Dhaka (BGS-DPHE).

[29] When examined more closely on the basis of the distribution of groundwater As, the separation between tube

wells constrained to reach Pleistocene sands below the Madhupur clay and tube wells tapping into Holocene aquifers does not seem to run uninterrupted even within the northwestern half of the study. Hand-drawn contours of wells that yield water with up to 50 μg/L As (Figure 7) outline a gap corresponding to two dozen higher-As tube wells within the 30–45 m (100–150 ft) depth interval and below 45 m (150 ft). Probably not by coincidence, the same area contains a large number of shallower wells in the 15–30 m (50–100 ft) range, a majority of which yield water containing over 50 μg/L As. This suggests a different depositional history than the low-As area immediately to the southwest with hardly any wells shallower than 30 m (100 ft). The stream running today through the part of the gap demarked by the contours of low-As wells suggests that a larger stream may have cut through the Madhupur Clay in the past and that this channel was subsequently filled with Holocene sediment, which resulted in higher groundwater As concentrations extending to greater depth (Figure 7).

4.5. Implications for Mitigation

[30] Even if imperfectly understood at this point, the spatial patterns in groundwater As documented for a portion of Araihazar at an unprecedented combination of coverage and resolution have a number of implications for mitigation. Within the village with a highly mixed As distribution, households with a well high in As more often than not live within walking distance of a neighboring well that is low in As (Figure 9). This sort of patchiness could clearly be exploited to reduce As exposure, at least in the short term, if social barriers to sharing the subset of safe but privately-owned wells could be overcome [*van Geen et al.*, 2002; Gelman et al., submitted manuscript, 2002]. Temporal changes are a source of concern, however, especially for the shallow wells that meet the local drinking water standard but are surrounded by wells that do not. We have so far monitored a half dozen such wells in Araihazar, both deep and shallow, over a year and observed no significant changes in response to the monsoon-driven changes in water table [*van Geen et al.*, 2002]. Relatively constant groundwater As concentrations have also been reported for a limited number of other well-documented time series (BGS-DPHE).

[31] Sharing existing wells is not a viable mitigation option in the second close-up area where most of the households live several hundred meters from the closest safe wells (Figure 10). We therefore chose this particularly affected region to install several community wells, guided by a nest of monitoring wells installed in the same village next to the coring site (Zheng et al., manuscript in preparation, 2003). The water supplied by three of the new community wells that were installed at 60 m (200 ft) depth, which we continue to monitor, also meets the Bangladesh standard for drinking water [*van Geen et al.*, 2003a]. However, only 1 km to the north and 1 km to the south of this region, two wells initially installed to a depth of 60 m (200 ft) produced water containing 115 and 94 μg/L As, respectively (Figure 7). The separation of deep aquifers low in As from shallower aquifers therefore appears to be poorly defined in terms of both the depth and the extent of the relatively thin clay layers in the second close-up area. By drilling deeper at the same two locations, using a hydraulic pump that is currently not available to the local drillers, another drilling team was eventually able to reach groundwater containing <5 μg/L at 120 m (400 ft) and 138 m (460 ft), respectively. The six community wells installed in the eastern portion of our study area have become remarkably popular. Many villagers walk hundreds of meters each day to fetch safe drinking water from the new community wells. A study of well usage has shown that, on average, 2200 L/day is hand-pumped from such a well regardless of the season, and that a single community well can meet the needs of about 500 people [*van Geen et al.*, 2003a]. To build on these encouraging results, optimal locations for the installation of additional community wells were recently determined from a spatial analysis of the As data for existing wells in our study area (Gelman et al., submitted manuscript, 2002).

[32] The local drillers with whom we have worked know that orange-brown sediment is associated with low-As groundwater. One of the drillers in the area appears to already have some spatial sense of how the depth to the orange-brown sediment varies from one village to the other. Systematic mapping of the depth of transition to these deposits every time a well is installed, based on cuttings, would clearly be beneficial for more systematic targeting low-As aquifers. Our failure to predict the depth to safe water in two out of a half-dozen community wells despite our mapping capabilities indicates that the task is likely to remain difficult, however. Even though the Dupi Tila sand formation appears to be accessible with local drilling technology based on the "hand flapper" method (maximum penetration of 90 m (300 ft)) at most locations within our study area, this method is not adequate in areas further south such as Lakshmipur where a penetration greater than 300 m (900 ft) is required (BGS-DPHE). Households might be willing to pay for the additional expense of hydraulic pumps to drill deeper wells in such areas, if the local drillers could provide that service.

[33] In contrast to the conclusions drawn by the BGS-DPHE team, we are not convinced that installation of tube wells within Holocene aquifers should be entirely banned. The distribution of As in tube wells less than 16 m (50 ft) deep in Araihazar indicates that at least in some villages, the likelihood of encountering shallow low-As groundwater is quite high. Again, mapping such characteristics and conveying this information to local drillers could be very beneficial. There are some other groundwater constituents whose concentration can exceed WHO guidelines values in shallow wells, however (BGS-DPHE). Although we have not yet conducted a full assessment, elevated Mn concentrations in shallow wells throughout Araihazar are a source of concern [*van Geen et al.*, 2003a].

[34] An alternative approach to the difficult task of mapping low-As aquifers is to determine during the drilling process itself whether an aquifer is likely to be elevated in As before a well is installed. This is a direction worth pursuing because it might allow local drillers to more effectively target low-As aquifers while reducing the cost of the installation to the household. This is because the sections of PVC pipe and other supplies used in the construction of a well account for approximately half the total cost in Bangladesh. Once a well has been installed and the As content of the groundwater is found to be too high, most of these

materials cannot be used again. As a potential aid to predicting groundwater As ahead of installation, we successfully tested an inexpensive sediment reflectance meter constructed by modifying a flashlight to determine the proportion of Fe III and Fe II oxyhydroxides in the sediment with two light-emitting diodes and a detector [*Kostel*, 2002]. We recently also demonstrated that a simple needle-based device mounted at the end of the drill rig used by the local teams can be used to sample representative groundwater ahead of the drill hole (A. van Geen, manuscript in preparation, 2003). Because the relation between sediment reflectance and groundwater As can be ambiguous (A. Horneman et al., manuscript in preparation, 2003), we envision that the reflectance meter could be used as a rapid screening tool during drilling and that the needle-sampler could be used for collecting and testing groundwater before the final installation of a well. Because of continuing concerns about the reliability of existing field kits for arsenic [*Rahman et al.*, 2002], any approach that takes advantage of the patchiness of the groundwater As to reinstall safe wells would greatly benefit from an improved field kit for As and its availability on demand at the local scale.

5. Conclusions

[35] About half the nearly 6000 tube wells sampled in Araihazar supply water with less than 50 μg/L As, while the other half do not meet the Bangladesh standard for drinking water. For reasons that are unclear at this point, the greatest number of wells in Araihazar extend to aquifers in the 12–24 m (40–80 ft) depth range where As concentrations most frequently exceed 50 μg/L. The spatial distribution of tube well As is consistent with the location of the study area at a geologic boundary. The oxic low-As aquifers accessible by drilling with local technology appear to be of Pleistocene age; the shallower reducing aquifers often containing elevated but variable As concentrations were deposited since the Last Glacial Maximum ~20,000 years ago.

[36] The various patterns of As distributions that we have documented for Araihazar appear to be representative of the different modes of variability that prevail in other parts of Bangladesh. There is considerable potential for exploiting these patterns to tackle the arsenic crisis in Araihazar and beyond. The high degree of patchiness of As in shallow Holocene aquifers of many villages indicates that, following a blanket survey, a short-term mitigation option is to encourage sharing of the wells that yield water containing less than 50 μg/L [*van Geen et al.*, 2002]. Although results from monitoring a half-dozen shallow low-As tube wells in Araihazar have been encouraging so far, a subset of such wells should be regularly monitored in an area with spatially highly-variable As levels in groundwater.

[37] Well-switching is not an option for a significant number of villages in Araihazar where essentially all existing wells tap into aquifers with over 50 μg/L As. In such areas, recently installed community wells that reach low-As Pleistocene aquifers have been remarkably popular. The depth of such aquifers varies from 30 m (90 ft) to over 130 m (390 ft) within a distance of a few kilometers within our study area, however. This does not appear to be the case only because Araihazar is geologically a transitional area. Pronounced differences of the depth of relatively oxic Pleistocene aquifers underline the value of combining As and depth information at the village-scale to facilitate targeting of low-As groundwater. The spatial variability of the depth of Pleistocene aquifers also indicates the futility of establishing a "safe" depth at the upazilla level, let alone the national level. Instead, government and NGOs should be urged to provide hand-held GPS receivers to the numerous village workers currently testing tube wells throughout the country. Combined with depth information that can be obtained from the owners of each well, such data obtained at little additional cost could be used to map the distribution of As and the associated geologic formations from the national to the village level.

Appendix A

[38] A simple model is needed to quantify the impact of the various sources of error on the As data. In this model, X_i is the estimate of the As concentration obtained from a single measurement relative to a calibration curve obtained from standards diluted in pure matrix modifier solution. X_i is assumed to be a linear function of the actual As concentration θ, some error $\varepsilon_{Xi,}$, and the ratio of the instrument response for samples diluted in matrix modifier solution relative to the response for a pure matrix, expressed by a factor $(1 + \varepsilon_{cal})$. This factor has a mean of 1 and a standard deviation σ_{cal} that needs to determined; ε_{Xi} are independent errors for each measurements with a mean of zero and a standard deviation of σ_{meas}.

$$X_i = (1 + \varepsilon_{cal})\theta_i + \varepsilon_{Xi}$$

[39] Three measurements enter into the measurement of the concentration increase that followed the addition of an As spike in the field. X_1 and X_2 are replicate measurements; X_3 is the measurement for the spiked sample. Although the concentration of this spike is known, there is some uncertainty in the resulting concentration increase in the sample because of some error ε_{dil} in control of both the sample volume and the spike volume.

$$X_1 = (1 + \varepsilon_{cal})\theta + \varepsilon_{X_1}$$

$$X_2 = (1 + \varepsilon_{cal})\theta + \varepsilon_{X_2}$$

$$X_3 = (1 + \varepsilon_{cal})(\theta + 499)(1 + \varepsilon_{dil}) + \varepsilon_{X_3}$$

[40] After rearranging, the difference Y in concentration between the spiked sample and the average of the two replicates becomes:

$$\begin{aligned} Y &= X_3 - (X_1 + X_2)/2 \\ &= (1 + \varepsilon_{cal})(499 + (\theta + 499)\varepsilon_{dil}) + \varepsilon_{X_3} - (\varepsilon_{X_1} + \varepsilon_{X_2})/2 \end{aligned}$$

The mean of Y is 499 μg/L; the standard deviation of Y can be approximated with Taylor series by rearranging and summing the squares of the relative standard deviations for errors due to variations in the sensitivity of the instrument to the sample matrix σ_{cal}, variations in dilutions σ_{dil}, and the reproducibility of sampling and analysis σ_{meas}.

$$\sigma_Y \simeq 499\sqrt{\left(\sigma_{cal}^2 + (1 + (\theta/499))^2\sigma_{dil}^2 + 3/2(\sigma_{meas}/499)^2\right)}$$

The single measurement error σ_{meas} = 7 μg/L is estimated in the quality control section from replicate analyses; the relative error corresponding to the spike of 499 μg/L is 0.014. Two factors need to be considered to determine σ_{dil}: the volume of sample added to the spike in the field that could not be controlled with the overflowing graduated cylinder to better than 60 ± 2 mL (a relative uncertainty of 0.033), and a smaller relative uncertainty of 0.01 in the 0.6 mL volume delivered by the two pipettes used for the As spikes. These errors in volume control add a total relative uncertainty to the expected increase in concentration σ_{dil} = 0.034. Since σ_Y = 56 μg/L is known from the 341 sets of replicates and spiked samples, we can estimate σ_{cal} from the expression above by substituting the mean As concentration for the (unspiked) replicates of 95 μg/L for θ. Solving yields σ_{cal} = 0.103, which confirms that the dependence of the instrument response on the sample matrix is probably the main factor that determines the total relative standard deviation for concentration differences between spiked and unspiked samples (0.112).

[41] From the values obtained for σ_{cal} and σ_{meas}, we can now estimate the absolute uncertainty of a single As measurement based on an instrument calibration with standards in pure matrix modifier solution:

$$\sigma_{sing} = \sqrt{\left(\theta^2\sigma_{cal} + \sigma^2_{meas}\right)}$$

According to this expression, the absolute uncertainty of 7 μg/L of obtained from replicate analyses dominates below 70 μg/L. Above this concentration, the relative error of ~10% due to differences in sample matrix becomes the larger source of uncertainty.

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References

Bangladesh Rural Advance Commission (BRAC), Combatting a deadly menace: Early experiences with a community-based arsenic mitigation project in Bangladesh, *Res. Monogr. Ser. 16*, Dhaka, Bangladesh, 2000.

Berg, M., H. C. Tran, T. C. Nguyen, H. V. Pham, R. Schertenleib, and W. Giger, Arsenic contamination of groundwater and drinking water in Vietnam: A human health threat, *Environ. Sci. Technol.*, *35*, 2621–2626, 2001.

Bhattacharya, P., G. Jacks, J. Jana, A. Sracek, J. P. Gustafsson, and D. Chatterjee, Geochemistry of the Holocene alluvial sediments of Bengal Delta Plain from West Bengal, India: Implications on arsenic contamination in groundwater, in *Groundwater Arsenic Contamination in the Bengal Delta Plain of Bangladesh*, edited by G. Jacks, P. Bhattacharya, and A. A. Khan, *TRITA-AMI Rep. 3084*, pp. 21–40, Kungl Tekniska Högskolan, Stockholm, 2001.

Biswas, B. K., R. K. Dhar, G. Samantha, B. K. Mandal, D. Chakraborti, I. Faruk, K. S. Islam, M. Chowdury, A. Islam, and S. Roy, Detailed study report of Samta, one of the arsenic-affected villages of Jessore District, *Bangladesh, Curr. Sci.*, *74*, 134–145, 1998.

British Geological Survey and Department of Public Health Engineering, *Arsenic Contamination of Groundwater in Bangladesh,* vol. 2, *Final Report*, *BGS Tech. Rep. WC/00/19*, edited by D. G. Kinniburgh and P. L. Smedley, Br. Geol. Surv., Keyworth, UK, 2001.

Burgess, W. G., M. Burren, J. Perrin, and K. M. Ahmed, Constraints on sustainable development of arsenic-bearing aquifers in southern Bangladesh. part 1: A conceptual model of arsenic in the aquifer, in Sustainable Groundwater Development, edited by K. M. Hiscock, M. O. Rivett, and R M. Davidson, *Geol. Soc. London Spec. Publ.*, *193*, 145–163, 2000.

Burgess, W. G., K. M. Ahmed, J. Cobbing, M. Cuthbert, S. Mather, E. McCarthy, and D. Chaterjee, Anticipating changes in arsenic concentration at tubewells in alluvial aquifers of the Bengal Basin, paper presented at Natural arsenic in sedimentary aquifers, XXXII IAH Congress on Groundwater and Human Development, Int. Assoc. for Hydrogeol., Mar del Plata, Argentina, 2002.

Chakarborty, A. K., and K. C. Saha, Arsenic dermatosis from tubewell water in west Bengal, *Indian J. Medical Res.*, *85*, 326–334, 1987.

Chen, Y., and H. Ahsan, Cancer burden due to arsenic exposure from drinking water in Bangladesh, *Am. J. Public Health*, in press, 2003.

Cornell, R. M., and U. Schwertmann, The iron oxides: Structure, properties, reactions, occurrences, and uses, VCH Verlagsgesellshaft, Weinheim, Germany, 1996.

Cummings, D. E., F. Caccavo, S. Fendorf, and R. F. Rosenzweig, Arsenic mobilization by the dissimilatory Fe(III)-reducing bacterium Shewanella alga BrY, *Environ. Sci. Technol.*, *33*, 723–729, 1999.

Datta, D. K., and V. Subramanian, Distribution and fractionation of heavy metals in the surface sediments of the Ganges-Brahmaputra-Meghna river system in the Bengal Basin, *Environ. Geol.*, *36*, 93–101, 1998.

Department of Public Health Engineering, Mott MacDonald Ltd, and British Geological Survey, Groundwater studies for arsenic contamination in Bangladesh, main report, Br. Geol. Surv., Keyworth, UK, 1999.

Dhar, R. K., et al., Groundwater arsenic calamity in Bangladesh, *Curr. Sci.*, *73*, 48–59, 1997.

Ferguson, J., On recent changes in the delta of the Ganges, *Q. J. Geol. Soc. London*, *19*, 321–354, 1863.

Goodbred, S. L., Jr., and S. A. Kuehl, Holocene and modern sediment budgets for the Ganges-Brahmaputra river system: Evidence for highstand dispersal to floodplain, shelf, and deep-sea depocenters, *Geology*, *27*, 559–562, 1999.

Goodbred, S. L., Jr., and S. A. Kuehl, The significance of large sediment supply, active tectonism, and eustasy on margin sequence development: Late Quaternary stratigraphy and evolution of the Ganges-Brahmaputra delta, *Sediment. Geol.*, *133*, 227–248, 2000.

Goodbred, S. L., Jr., S. A. Kuehl, M. S. Steckler, and M. H. Sarker, Controls on facies distribution and stratigraphic preservation in the Ganges-Brahmaputra delta sequence, *Sediment. Geol.*, *155*, 301–316, 2003.

Harvey, C. F., et al., Arsenic mobility and groundwater extraction in Bangladesh, *Science*, *298*, 1602–1606, 2002.

Kostel, K., A simple sediment-color meter to target low-arsenic aquifers in Bangladesh, M.S. thesis, Columbia Univ., New York, 2002.

Lovley, D. R., Magnetite formation during microbial dissimilatory iron reduction, in *Iron Biominerals*, edited by R. B. Frankel and R. P. Blakemore, pp. 151–166, Plenum, New York, 1990.

McArthur, J. M., P. Ravenscroft, S. Safiullah, and M. F. Thirlwall, Arsenic in groundwater: Testing pollution mechanisms for sedimentary aquifer in Bangladesh, *Water Resour. Res.*, *37*, 109–117, 2001.

Nickson, R., J. McArthur, W. Burgess, K. M. Ahmed, P. Ravenscroft, and M. Rahman, Arsenic poisoning of Bangladesh groundwater, *Nature*, *395*, 338, 1998.

Nickson, R. T., J. M. McArthur, P. Ravenscroft, W. G. Burgess, and K. M. Ahmed, Mechanism of arsenic release to groundwater, Bangladesh and west Bengal, *Appl. Geochem.*, *15*, 403–413, 2000.

Rahman, M. M., et al., Effectiveness and reliability of arsenic field testing kits: Are the million dollar screening projects effective or not?, *Environ. Sci. Technol.*, *36*, 5385–5394, 2002.

Smith, A. H., E. O. Lingas, and M. Rahman, Contamination of drinking-water by arsenic in Bangladesh: A public health emergency, *Bull. World Health Organ.*, *78*, 1093–1103, 2000.

Solley, W. B., R. R. Pierce, and H. A. Perlman, Estimated use of water in the United States in 1995, *U.S. Geol. Surv.*, *1200*, 1998.

Umitsu, M., Late Quaternary sedimentary environments and landforms in the Ganges Delta, *Sediment. Geol.*, *83*, 177–186, 1993.

van Geen, A., et al., Promotion of well-switching to mitigate the current arsenic crisis in Bangladesh, *Bull. World Health Organ.*, *80*, 732–737, 2002.

van Geen, A., K. M. Ahmed, A. A. Seddique, and M. Shamsudduha, Community wells to mitigate the current arsenic crisis in Bangladesh, *Bull. World Health Organ.*, in press, 2003a.

van Geen, A., Y. Zheng, M. Stute, and K. M. Ahmed, Comment on "Groundwater arsenic and irrigation in Bangladesh" (II), *Science*, *300*, 584, 2003b.

World Bank, *World Development Report 1998/99*, Oxford Univ. Press, New York, 1999.

World Health Organization, *Guidelines for Drinking-Water Quality*, vol. 2, *Health Criteria and other Supporting Information*, 2nd ed., pp. 940–949, Geneva, Switzerland, 1996.

Yu, W. H., C. M. Harvey, and C. F. Harvey, Arsenic in groundwater in Bangladesh: A geostatistical and epidemiological framework for evaluating health effects and potential remedies, *Water Resour. Res.*, *39*, doi:10.1029/2002WR001327, in press, 2003.

Zheng, Y., M. Stute, A. van Geen, I. Gavrieli, R. Dhar, H. J. Simpson, P. Schlosser, and K. M. Ahmed, Redox control of arsenic mobilization in Bangladesh groundwater, *Appl. Geochem.*, in press, 2003.

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