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Statistical characterization of arsenic contamination in shallow tube wells of western Bangladesh

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Abstract:

A regional assessment of the arsenic (As) contamination scenario in shallow tube wells (depth <150 m) of western Bangladesh is presented. Comparisons are made in light of bulk geological differences (Pleistocene versus Holocene deposits/northwest versus southwest) and As measurement protocols (field kit (FK) versus atomic absorption spectroscopy (AAS)). Our As database comprised the following: (1) the nationwide As survey completed in 1999 by the British Geological Survey in collaboration with the Department of Public Health Engineering (DPHE); and (2) a regional As survey conducted in southwest Bangladesh by the Japan International Cooperation Agency in collaboration with DPHE in 2002. First, we characterize the error structure of the semi-quantitative FK As measurements using collocated AAS As measurements as reference from a set of 307 wells located in southwest Bangladesh. The depth distribution of As is identified using a very dense cluster of 2963 wells over a 560 km^2 domain. The probability of the FK method for successful detection of a well sample as unsafe (safe) was found to be 96·9% (34·1%) and 95·2% (80·3%) for the World Health Organization (WHO) and Bangladesh safe limits, respectively. Similarly, the probability of false alarms and false hopes for WHO (Bangladesh) safe limits were found to be 3·1% (4·8%) and 87·5% (19·7%), respectively. The depth at which the highest fraction of wells exceeding a given safe limit occurred could still be inferred correctly by FK measurements. A simple bias adjustment procedure on FK As data did not result in a more accurate characterization of depth distribution of As. This indicated that simple error statistics are inadequate for advancing the utility of FKs; rather, an understanding of the complex and multidimensional error structure is required. Regional anisotropy in the spatial dependence of As for the northwest was found to be stronger than the southwest. The correlation length for As concentration in the east–west direction of northwest Bangladesh (i.e. across major river floodplains) was found to be almost twice (158·80 km) that of the north–south direction (along the major axis of Pleistocene deposits) (78·21 km). For the southwest region, the ratio of east–west to north–south correlation lengths ranged from 1·40 to 1·51. For the northwest region, because it is well known to have the lowest concentrations of As countrywide, knowledge of this anisotropy appears to suggest the need for drilling twice as many remediation deep wells in the proximity of an unsafe shallow well in the north–south direction than in the east–west direction. Findings from this study are potentially useful in setting priority areas for emergency testing, distributing remediation resources equitably and formulating a regional water resources strategy for western Bangladesh. Copyright © 2005 John Wiley & Sons, Ltd.

KEY WORDS arsenic contamination; groundwater; western Bangladesh; spatial patterns; field-kits; atomic absorption spectroscopy; remediation

INTRODUCTION

The water supply in Bangladesh is based primarily on groundwater. Estimates show that about 103 million (80% of the population) depend on shallow tube wells that have been excavated to a depth of less than 150 m

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(hereafter called 'shallow wells') (Ahmed, 2002, 2003). However, large-scale contamination of drinking water by arsenic (As) has been reported. Since 1993, when it was discovered that the alluvial Ganges aquifers of Bangladesh were contaminated with As, numerous studies and surveys have been conducted to understand the contamination scenario better. In the first nationwide survey of As, it was revealed that about 27% of the shallow wells exceeded the Bangladesh safe limit of 50 $\mu g\ l^{-1}$ (or parts per billion, ppb) while 46% exceeded the World Health Organization (WHO) limit of 10 ppb (Kinnburgh and Smedley, 2001). Figures for 'deep' wells (depth >150 m) were, however, found to be less alarming, being 1% and 5%, respectively. A summary of past studies and surveys can be found in Yu *et al.* (2003), van Geen *et al.* (2003) and McArthur *et al.* (2001), among others. Given the present state of the art, it is estimated that more than half of the population in Bangladesh is at risk from As poisoning.

It is generally accepted that As in groundwater is geologic in origin, deriving from the sediments from the upland Himalayan catchments (Kinnburgh and Smedley, 2001; McArthur *et al.*, 2001; Yu *et al.*, 2003). The study by the British Geological Survey (BGS) in collaboration with the Bangladesh Department of Public Health Engineering (DPHE) (Kinnburgh and Smedley, 2001) clearly demonstrated that, contrary to local knowledge of the villagers, there indeed exist spatial patterns of As contamination on the regional scale, that is the 50–100 km scale at which geology varies in Bangladesh (see Figure 1). The association of low levels in groundwater As is found with relatively oxic, uplifted old Pleistocene aquifers, and high As concentrations in reducing young Holocene aquifers (Nickson *et al.*, 1998). Most of the Pleistocene deposits are located in the northern region (comprising Barind clay and alluvial fan deposits), whereas the young Holocene deposits are located in the floodplains in the south (deltaic deposits and alluvial deposits) (see Figure 1; Alam *et al.*, 1990). It is, therefore, no surprise that the lowest and highest As concentrations are found in the northern and southern regions, respectively.

A very important scientific question is thus: What are the implications of this regional As spatial dependence in the context of regional-scale remediation efforts for shallow wells? Yu *et al.* (2003) estimated that, by replacing 31% of the country's shallow wells with deep ones, Bangladesh could potentially reduce the As-related health hazards by about 70%. Therefore, a statistical characterization of the spatial dependence of As at the regional scale can aid in the following: (1) establishing priority areas for emergency testing; (2) assisting in equitable distribution of remediation resources for deep well drilling; and (3) formulating a regional water resources strategy. Among the numerous studies conducted on As contamination of groundwater in Bangladesh (e.g. Biswas *et al.*, 1998; Burgess *et al.*, 2000; Bhattacharya *et al.*, 2002; Harvey *et al.*, 2002; Mukherjee and Bhattacharya, 2002; Meharg and Rahman, 2003; van Geen *et al.*, 2003; Yu *et al.*, 2003), there are few that have characterized the spatial dependence of As contamination in light of bulk regional geological differences. This study, therefore, is motivated by the need to characterize the As contamination in western Bangladesh statistically as a function of regional geology. Western Bangladesh is defined as the region west of the two major river systems, the Brahmaputra and Meghna (see Figure 2), comprising about 50% of the country's population. It is a region that has been sparsely studied in light of As contamination because of the following possible reasons: (1) the lowest As concentrations (below the Bangladesh safe limit) are usually found in the northwest Pleistocene deposits; (2) the highest As concentrations, and hence most regional and local study areas, are concentrated in the south-central area of the country east of the Brahmaputra–Meghna river system (see Figure 2).

However, a major hurdle exists to such a statistical study targeting the characterization of regional patterns. The majority of As measurements on a large-scale regional basis (totaling about 1·3 million wells) are available from cost-effective but semi-quantitative field kits (FKs; DPHE-UNICEF, 2001; Rahman *et al.*, 2002). There are currently at least four different brands of FK in use in Bangladesh. Most of these kits used to date are based on the subjective mercuric bromide stain method, which has been reported to produce quantitatively meaningless results at As concentrations below 150 ppb (Abernathy *et al.*, 1997). The wisdom of the large-scale use of FKs has also been questioned recently (Rahman *et al.*, 2002). Thus, the most reliable As data available thus far are measurements by atomic absorption spectroscopy (AAS), which are available only on a limited scale due to the significantly higher operating costs. Therefore, it is also worthwhile characterizing

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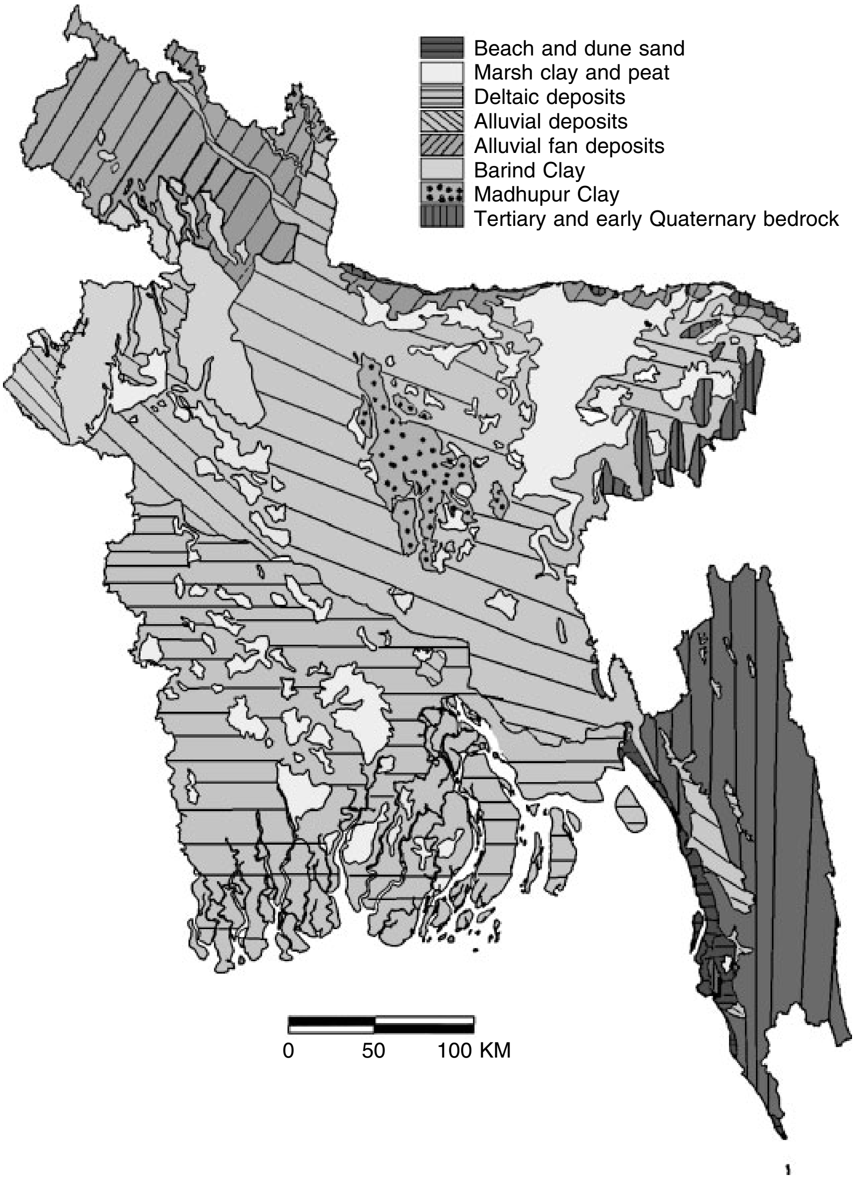

Figure 1. Geology of Bangladesh, based on Alam *et al.* (1990)

the error structure of the more widely used FKs for As measurement. In fact, Kinnburgh and Smedley (2001) recommends reliable FK analysis as one of the most important research directions for As remediation in Bangladesh. For more details on the specific brands and current research activity on field kits in Bangladesh, the reader is referred to the NGOs Arsenic Information and Support Unit (www.naisu.info).

The objectives of this study are twofold: (1) to identify the error structure of FK As measurements and explore the efficacy of simple statistical correction techniques; and (2) to characterize the spatial dependence of As concentration in shallow wells of western Bangladesh as a function of bulk geological structure. Results from this study are expected to provide preliminary insights on the usefulness of large-scale FK As estimates and possibly indicate ways towards advancing their use. More specifically, this study hopes to provide an

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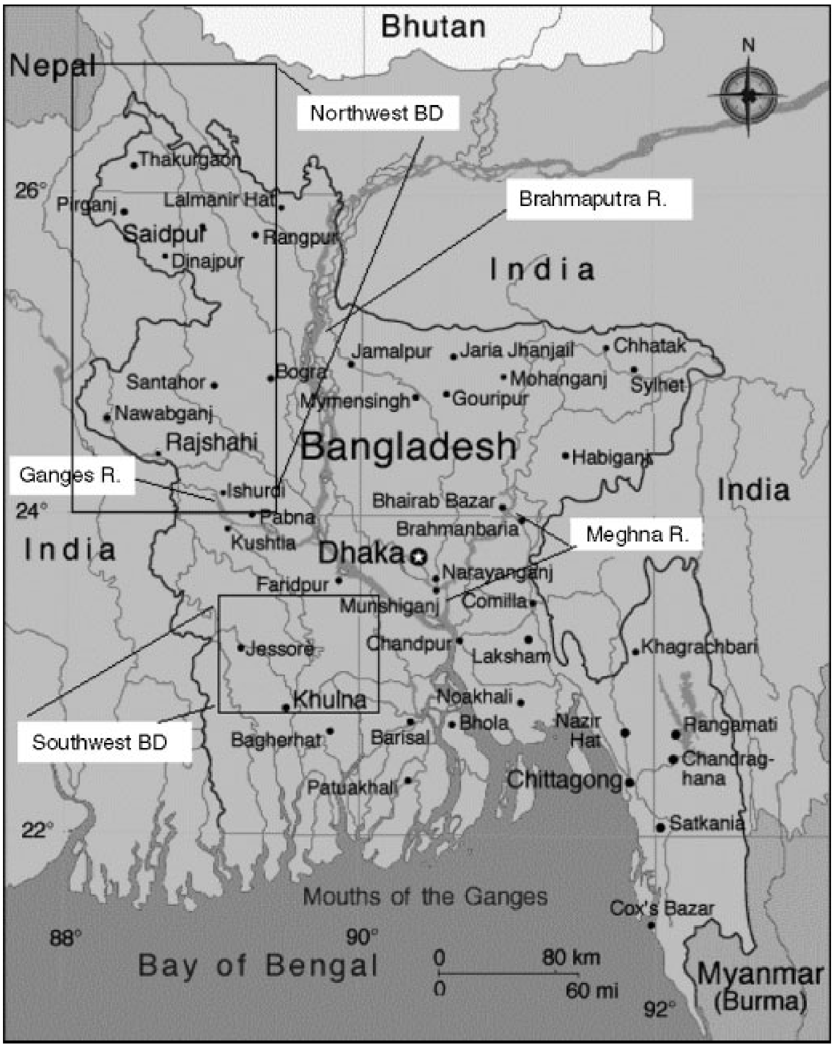

Figure 2. Study regions comprising northwest and southwest Bangladesh (upper and lower box respectively). The region west of Brahmaputra River and Meghna River constitutes western Bangladesh

objective framework to improve the problem definition for scientific and management questions, such as: Where should the priority areas for emergency testing be planned? How should the network for sinking deep wells be arranged? What guidelines need to be formulated for a regional water resources strategy in Western Bangladesh? Although it is recognized that there may be other important criteria that are not examined here, we hope that this paper will lead to a plethora of studies involving a wide range of measurement protocols, sampling methods, resolutions, geostatistical techniques and objectives towards an optimal strategy for regional-scale mitigation of arsenic contamination.

STUDY REGION AND DATA

We first define western Bangladesh as the region west of the Brahmaputra–Meghna river system (see Figure 2). The region is geographically divided into two parts based on geologic differences: (a) northwest Bangladesh, comprising mainly old oxic Pleistocene deposits (Figure 2, upper box); and (b) southwest Bangladesh, comprising mainly new Holocene deposits (Figure 2, lower box). Hereafter these regions are referred to as northwest BD and southwest BD, respectively. Based on As data from the Kinnburgh and Smedley (2001) field campaign (discussed next) and knowledge of the geology (Alam *et al.*, 1990), we defined northwest BD as the region bound by 24·0–26·7 °N latitude and 88·0–89·5 °E longitude, representing an area of about 35 000 km^2 (Figure 2). The region outside Bangladesh was not included in the analysis, due to the paucity of data. Based on recent As data availability from the JICA–DPHE (2002) field campaign (discussed next), southwest BD was bound by 22·49–23·79 °N latitude and 89·0–90·0 °E longitude, representing an exclusively Holocene area of about 13 000 km^2. A point to note is that the boundaries of the regions (northwest and southwest) were carefully selected so that they would reflect a predominant geologic character in the bulk sense. For example, the 89·5 °N cutoff for northwest BD avoids the floodplain Holocene regions of the Brahmaputra River. Additionally, the smaller scale sub-regional geologic variability (at the 5 km scale) was assumed to have a negligible effect on the overall make-up of regional geology. As an example, the small town (<25 km^2) of Chapai Nawabganj (see upper left portion of Figure 2) is known to possess a predominantly Holocene (alluvial deposits with high As) character surrounded overwhelmingly by Pleistocene geology (i.e. Barind clay with low As; see Figure 1). Hence, the average As concentration of 6 ppb for the whole Nawabganj district (>25 km^2) is observed by the BGS–DPHE survey to be very similar to a wholly Pleistocene counterpart district such as Jaipurhat (see Figure 4).

The As data pertained to the following two field campaigns:

- *Campaign 1*. The BGS–DPHE countrywide As survey was conducted from 1998 to 1999. This dataset comprises 3534 wells, with 850 of them being shallow wells in northwest BD. The dataset is available at http://www.bgs.ac.uk/arsenic/Bangladesh. Sample wells were systematically selected uniformly with approximately one well per 37 km^2. As with any type of field investigation, certain limitations (such as inaccessibility of sampling locations and local lack of familiarity with randomized sampling) existed with this survey. However, in the overall scheme of our investigations, such limitations are considered insignificant due to the fact that the BGS–DPHE survey currently represents the most quality-controlled database of As measurements available countrywide. A further point to note is that the As measurements of BGS–DPHE are based on the AAS method. For more details on this dataset, sampling and measurement protocols can be found in Kinnburgh and Smedley (2001: vol. 1).
- *Campaign 2*. The JICA–DPHE regional As survey was conducted by the Japanese International Cooperation Agency (JICA) in collaboration with DPHE in southwestern Bangladesh in 2002. This dataset has become available as part of a study on groundwater development of deep aquifers in southwest BD. This campaign had two components: (A) a small-scale study over a 560 km^2 area representing 2963 wells measured by the FK method with 300 collocated AAS measurements; (B) a large-scale study over a 13 000 km^2 area (referred earlier as southwest BD), which is inclusive of (A) and represents 307 shallow wells measured by both AAS and FK methods. The FK brand used was the Asia Arsenic Network (AAN) type and was imported from Japan. The merit of using an FK was to be able to know the approximate As concentration at the site in a very short time with minimum cost. A typical FK test usually costs about US$0·5 and takes less than 30 min to complete (Rahman *et al.*, 2002; www.naisu.info). The AAN FK method of As measurement followed the Hironaka method, which is considered to be a relatively robust semi-quantitative technique (Tanabe *et al.*, 1997). For the AAS method, an arsenic analysis laboratory was set up at the DPHE Jhenaidah office in 2000 (see bottom left portion of Figure 4 for location). The laboratory was equipped with an AAS unit having a continuous hydride vapour generator and an auto-sampler. The laboratory was also fitted with a water distillation unit and ion-exchange filter for pure water production. The testing capacity of the laboratory was 1000 samples per month. The locations of wells were measured by hand-held global positioning system devices that had an accuracy of about 5 m. Further details on Campaign 2 may be found in JICA–DPHE (2002). Owing to the high density of FK measurements for region (A) and the expected more modern sampling and measurement protocols that were followed (for both the FK and AAS methods),

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the As dataset for Campaign 2 is considered relatively unique and, to the best of our knowledge, has never been discussed in the current literature on As contamination of Bangladesh groundwater. Although certain limitations may be endemic in the JICA–DPHE survey (similar to those reported by Kinnburgh and Smedley (2001)), we have considered them consistently insignificant in the greater spirit of this study.

ERROR STRUCTURE OF FK MEASUREMENTS

The primary motivation for quantification of the error structure of FK measurements was the need to explore tangible ways to advance their usage given their low cost and the large size of their database (about 1·3 million wells). Past studies, such as Rahman *et al.* (2002), have concentrated on this issue, but without explicit demonstration of ways to minimize FK measurement limitations. The characterization of error structure for the FK As measurements was identified over 307 collocated AAS measurements of region (B) of Campaign 2 (JICA–DPHE, 2002). We assumed an As concentration of 1 ppb (1·0 $\mu g\ l^{-1}$) as the detection limit for current AAS measurement technology. Hence, the wells containing As concentration below this detection limit were assigned a value of 1 ppb. Assuming that As measurements by the AAS method (As_{AAS}) represented the most accurate concentration level, an FK As measurement (As_{FK}) may exhibit one of four possible outcomes during a sample test:

1. The FK method predicts a well to be safe when the As_{AAS} is less than the safe limit (i.e. successful safe well detection).
2. The FK method predicts a well to be safe when the As_{AAS} is greater than the safe limit (i.e. false hope).
3. The FK method predicts a well to be unsafe when the As_{AAS} is less than the safe limit (i.e. false alarm).
4. The FK method predicts a well to be unsafe when the As_{AAS} is greater than the safe limit (i.e. successful unsafe well detection).

Because our concern is primarily with the detection of unsafe wells and their potential remediation, we use outcome (4) as the conditional FK As prediction where the error in prediction ε is defined as

$$\varepsilon = \frac{As_{FK}}{As_{AAS}} \tag{1}$$

A point to note here is that the safe limit is assumed to be the AAS detection limit of 1 ppb for Equation (1). We also define the probabilities of detection (safe and unsafe), false alarm and false hope as follows:

$$\text{Probability of Detection of Safe Well} = \text{POD}_{\text{SAFE}}(As_{FK} < \text{limit} \mid As_{AAS} < \text{limit}) \tag{2}$$

$$\text{Probability of Detection of Unsafe Well} = \text{POD}_{\text{UNSAFE}}(As_{FK} > \text{limit} \mid As_{AAS} > \text{limit}) \tag{3}$$

$$\text{Probability of False Hope} = P_{\text{FalseHope}}(As_{FK} < \text{limit} \mid As_{AAS} > \text{limit}) \tag{4}$$

$$\text{Probability of False Alarm} = P_{\text{FalseAlarm}}(As_{FK} > \text{limit} \mid As_{AAS} < \text{limit}) \tag{5}$$

where limit represents the safe As limit (for either WHO or Bangladesh).

Table I provides a summary of the error structure of FK As measurements for two limits: WHO safe limit (10 ppb) and Bangladesh safe limit (50 ppb). We observe that the FK method has insignificant false alarm probability, but it can have significant probability for false hope ranging from 19·7% to 87·5% for the WHO and Bangladesh safe limits, respectively. This finding is in contrast to the FK error characterization by Rahman *et al.* (2002), where the probabilities for false alarm and false hope were both found to be greater than 30%. We hypothesize that this contrast is due to the use of the Hironaka method, which can be considered different from the semi-quantitative techniques used in other brands investigated by Rahman *et al.* (2002). Because all FK brands rely heavily on the operator's perception of the colour of stain produced

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Table I. Error structure of FK As measurements

	WHO safe limit (10 ppb) (%)	Bangladesh safe limit (50 ppb) (%)
POD_{SAFE}	12·5	80·3
$\text{POD}_{\text{UNSAFE}}$	96·9	95·2
$P_{\text{FalseAlarm}}$	3·1	4·8
$P_{\text{FalseHope}}$	87·5	19·7
Mean ε ($1/\lambda$)	1·24	

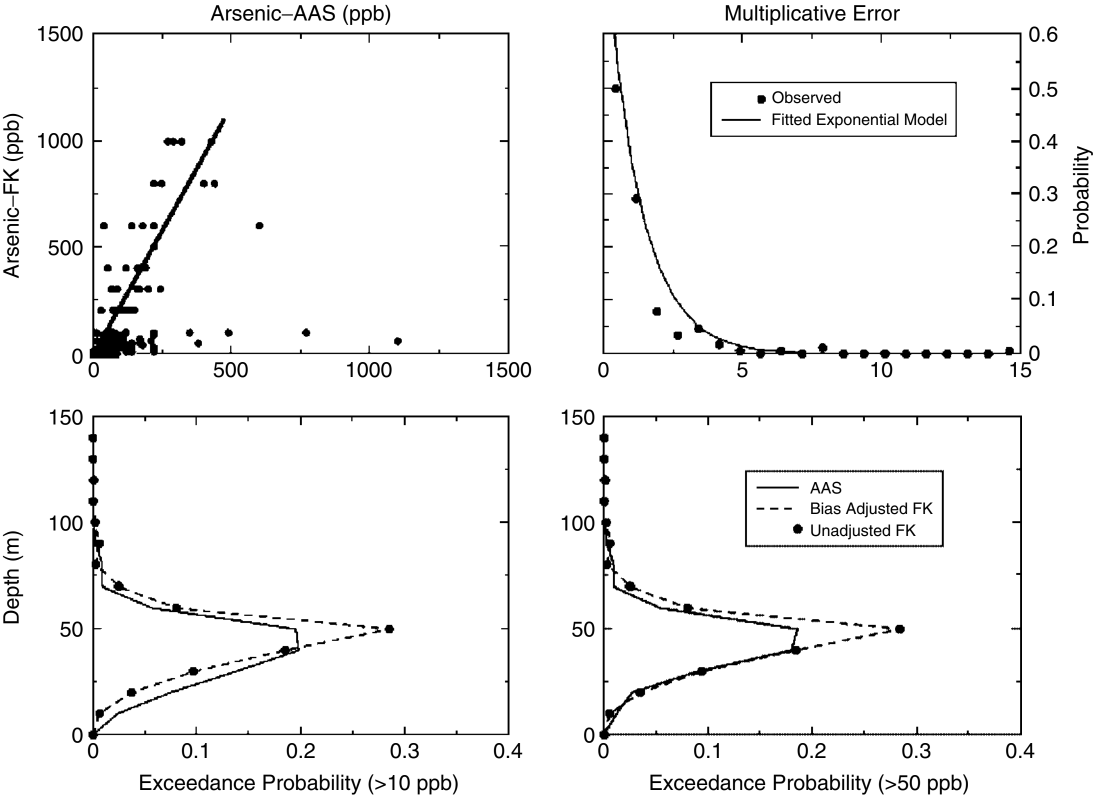

Figure 3. Error structure of FK As measurements. Upper left panel: scatter plot of FK As versus AAS As measurements; upper right panel: probability distribution of multiplicative error (conditional) for FK method; lower left panel: depth distribution of As concentration exceeding the WHO safe limit of 10 ppb; lower right panel: depth distribution of As concentration exceeding the Bangladesh safe limit of 50 ppb

during the test, a certain subjectivity remains. This thereby raises the probability of false hope and false alarms. However, organoleptic tests of 77 people of the students of Miyazaki University (Japan) indicated that the Hironaka method was insignificantly sensitive to an individual's perception of colour (Tanabe *et al.*, 1997).

The distribution of ε was found to be highly exponential, with a mean of 1·24 and standard deviation of 1·66 (Figure 3, upper right panel). The overall positive bias (1·24) is reconciled with the insignificant false alarm rate from the observation that the FK As results tend to be underestimated significantly at AAS concentrations below 44 ppb and overestimated above it (see next paragraph). The parameter λ for the exponential probability distribution ($P(x) = \lambda\ e^{-\lambda x}$) was therefore 0·806 (inverse of the mean). Because the exponential distribution

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has a mode of zero, the implication for the FK method is that it may have a high tendency not to detect As concentrations unless exceeding a certain threshold. This suggestion echoes previous studies, such as those of Rahman *et al.* (2002) and BRAC (2000), where the FK method was usually found inaccurate for AAS As concentrations below 100 ppb with a high degree of false hopes. In Figure 3, upper left panel, we show a scatter plot of $\mathrm{As}_{\mathrm{FK}}$ (y-axis) versus $\mathrm{As}_{\mathrm{AAS}}$ (x-axis), where the positive bias of the FK method is clearly observed.

As part of testing the effectiveness of a simple bias adjustment procedure on FK As measurements, we decomposed the mean of the error ε for the four important ranges (shown in parentheses): (1) 1–10 ppb (0·489); (2) >10–50 ppb (0·7176); (3) >50–100 ppb (1·194); and (4) >100–1000 ppb (3·14). A variable bias adjustment was then performed on an independent set of FK As data (comprising 2963 wells) for each of these ranges. The validation data pertained to region (B) of Campaign 2 (discussed above). A point to note is that the variable bias adjustment was performed only on wells that were considered detectable by the FK method (i.e. $\mathrm{As}_{\mathrm{FK}} > 1$ ppb) and, hence, represented a very simplified correction strategy that ignored the probability of false hopes. Because region (B) represented a dense cluster of wells, we assumed that the As data constitute a pseudo-point-like measurement due to the very small average intra-well distance. Hence, this dataset was considered suitable for characterizing the variation of As concentration with depth. We then developed the distribution of As exceeding a safe limit (i.e. fraction of wells unsafe) as a function of depth. The true depth As profile was identified from the 300 collocated As measurements by the AAS method for region (B). Comparisons were made between the bias-adjusted and -unadjusted FK As depth profiles with the true profile. The lower panels of Figure 3 show the effect of bias adjustment. We observe the following: (1) a simple variable bias adjustment has an insignificant effect on changing the depth distribution of As; (2) the FK As measurements, despite their complex error structure, can still adequately characterize the modal depth (depth of highest As contamination) for shallow wells. This probably indicates that the database of 1·3 million FK As measurements that are currently available countrywide may indeed be sufficient to characterize the probable depth of highest As contamination for local regions, even though the modal As contamination rate (i.e. 'highest fraction of contaminated wells') may have an error of about 50% (Figure 3, lower right panel). This partial utility of the FK method may, therefore, allow villagers to be aware of the risk of contamination for a shallow well provided that its depth is known accurately by the owner. Furthermore, the wells that are deemed 'probably contaminated' in this fashion by the local FK As depth distribution profile are, in fact, the ones that should be tested on a priority basis by the AAS method. In essence, the FK test can, therefore, act as a fast-running and inexpensive proxy to time-consuming and costly field campaigns based on AAS testing and, thereby, optimize resources. Considerable savings in operating expenditures can be expected by adopting a strategy as follows: (1) first, identify the modal depth of As distribution at the village level using blanket screening by FKs (a typical FK testing of 500 shallow wells at a village costs less than US$500 with a one-time capital cost of US$20–50; for details refer to www.naisu.info); (2) next, for definitive identification of unsafe wells, send selective samples of *only those wells corresponding to the FK-modal depth* to the nearest laboratory for the costly AAS testing. The potential use of the FK is, therefore, in helping to eliminate unnecessary testing of potentially safe wells, which should be considered less of a priority than the functional unsafe wells.

Finally, in the spirit of advancing the usage of FKs, the quantified parameters of the error structure allow a rational way of developing a potential FK error model. The utility of the FK error model will then lie in its ability to mimic hypothetical FK measurements by stochastically corrupting true AAS measurements. The successful detection ability of safe/unsafe wells by an FK can be modelled probabilistically using Bernoulli trials according to a scheme analogous to the simulation methodology for satellite rainfall detection recently developed by Hossain and Anagnostou (2004). Subsequently, the FK error model can then be used to understand the optimal performance criterion for effective countrywide management of As contamination using a finite set of AAS measurements as a benchmark.

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SPATIAL DEPENDENCE OF ARSENIC CONTAMINATION

The spatial dependence of As contamination was studied by means of variography analysis for northwest and southwest BD (i.e. region (A) of Campaign 2). We first regionalized the As concentration of wells by averaging over a 5 km × 5 km grid. Then the logarithm (base 10) of the averaged As values was taken. The assumption behind such a regionalization was that there exists little spatial structure of As at scales less than 5 km (Kinnburgh and Smedley, 2001; Yu *et al.*, 2003) and also that there currently exists no reliable countrywide As database to explore spatial variability at finer scales. For the northwest BD, the As data pertaining to Campaign 1 (Kinnburgh and Smedley, 2001) were used. For southwest BD, data from both Campaigns 1 and 2 (JICA–DPHE, 2002) were used. Because the dataset of Campaign 2 is considered unique and very recent to the current literature, comparisons of spatial dependence analyses with Campaign 1 could possibly reveal a refinement of the picture on As variability at a regional scale. Anticipation of a refinement is justified because of the following: (1) Campaign 2 had prior knowledge of findings from Campaign 1 and, hence, sampled wells *a posteriori*; (2) Campaign 2 employed a more up-to-date set of As measurements and data management protocols than Campaign 1 (as discussed in the 'Study region and data' section). Henceforth, all subsequent analyses and discussions shall refer to Campaigns 1 and 2 as BGS–DPHE and JICA–DPHE respectively.

The variogram, which represents the variation between pairs of As measurements as a function of separation distance, was calculated as follows:

$$\gamma(h) = \frac{1}{2n(h)} \sum_{i=1}^{n(h)} [z(x_i) - z(x_i + h)]^2 \tag{6}$$

where $z(x_i)$ and $z(x_i + h)$ are the regionalized logarithms of regionalized As values at x_i and $x_i + h$ respectively and h (km) is the lag. The term $\gamma(h)$ shall hereafter be referred to as simply 'variance'. Note that, because As measurements were regionalized on a 5 km × 5 km grid, the variogram in Equation (6) was computed every 5 km (up to a maximum lag of 50 km) with the zero lag effectively being at 5 km. All As measurements pertained to the AAS method. We first assumed isotropy in the As variation for both northwest and southwest BD and computed the isotropic (omnidirectional) variogram. Next, assuming that anisotropy existed due to geologic conditions, we computed the anisotropic (directional) variogram for two specific directions, i.e. north–south and east–west. The choice of these two orthogonal directions is justified as follows. For northwest BD, the north–south strips of deposits are mainly Pleistocene deposits and located away from the relatively new sediment-laden floodplains in the east–west direction (see Figures 1 and 2). For southwest BD, the region is more geologically isotropic than northwest BD. However, the tendency of increasing As concentrations towards the south-central region (east of Meghna River, Figure 2) imposes some amount of anisotropy in the As variability. Reference to the map for district-mean As concentration (ppb) derived from Kinnburgh and Smedley (2001) clarifies the case for anisotropy further (Figure 4). One can clearly observe that the variability of As concentrations is not the same for north–south and east–west directions, and this holds true for both northwest and southwest BD.

A quantitative analysis of a variogram derived as described above is performed by fitting a variogram model. Assuming that the variogram is best represented by an exponential model, we fitted the functional parameters describing the spatial variability as follows:

$$\gamma(h) = c_0 + c(1 - \mathrm{e}^{-h/a}) \tag{7}$$

where c_0 represents the nugget variance, c is the sill variance and a is the distance parameter, often known as 'correlation length'. The correlation length is analogous to the rate at which spatial dependence decays as a function of separating distance. Model fitting was performed by the method of least squares, where we assumed the lag-5 km variance $\gamma(5)$ as nugget variance c_0 and the lag-50 km variance $\gamma(50)$ as the sill variance c. This may be considered as a gross approximation, but the purpose of the model fitting exercise

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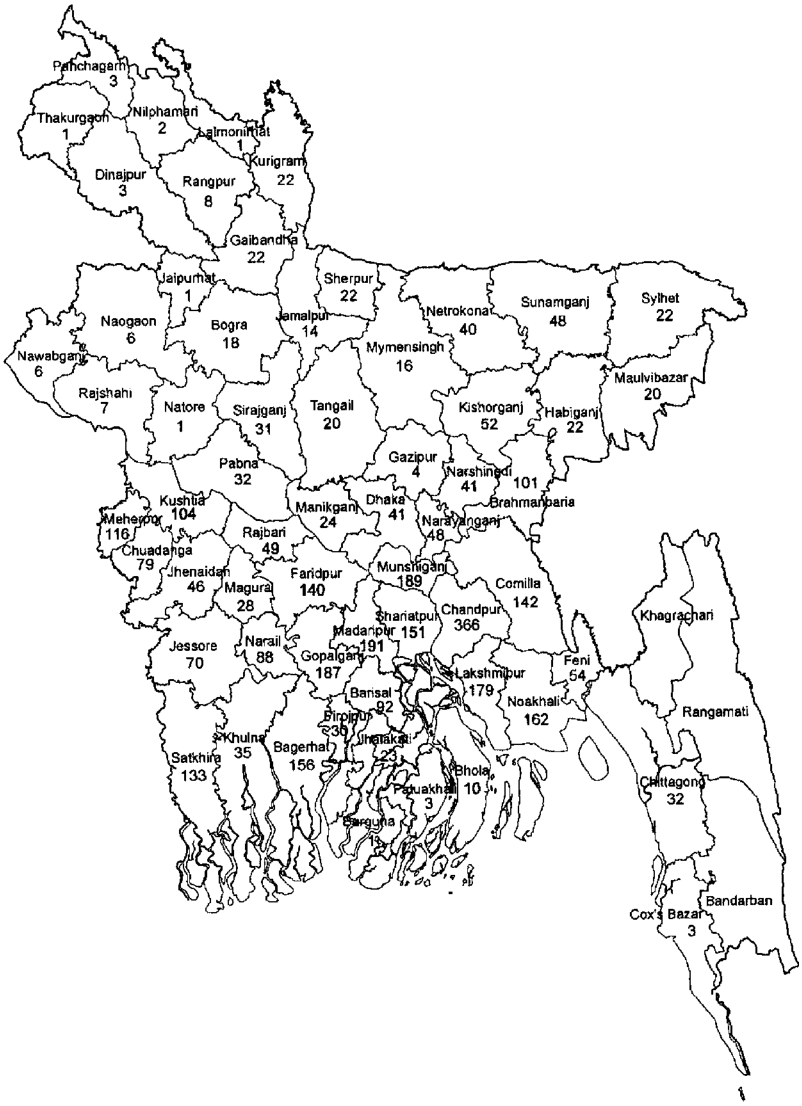

Figure 4. Map of the district-mean As concentrations (ppb) based on the Kinnburgh and Smedley (2001) survey

was to determine a parameter (the correlation length *a*) for quantifying and comparing the spatial dependence. We believe that the choice of nugget and sill variance will have a negligible effect on the calibration for correlation length in the 5–50 km length scales.

Figure 5 presents the isotropic variogram for the northwest and southwest BD for the BGS–DPHE and JICA–DPHE campaigns. Under the assumptions of isotropy, the spatial variability of As is found to be considerably less in northwest BD than in southwest BD. The southwest variograms computed by BGS–DPHE and JICA–DPHE appear very similar. However, in terms of correlation lengths, they represent a slightly different picture. The correlation length for northwest BD (by BGS–DPHE) is found to be 87·21 km, and for southwest BD it was 113·51 km. For JICA–DPHE, we calculated the correlation length to be 83·61 km (see Table II). The difference in correlation lengths for the BGS–DPHE and JICA–DPHE surveys in southwest BD can probably be attributed to the newer As measurement and data management protocols that have

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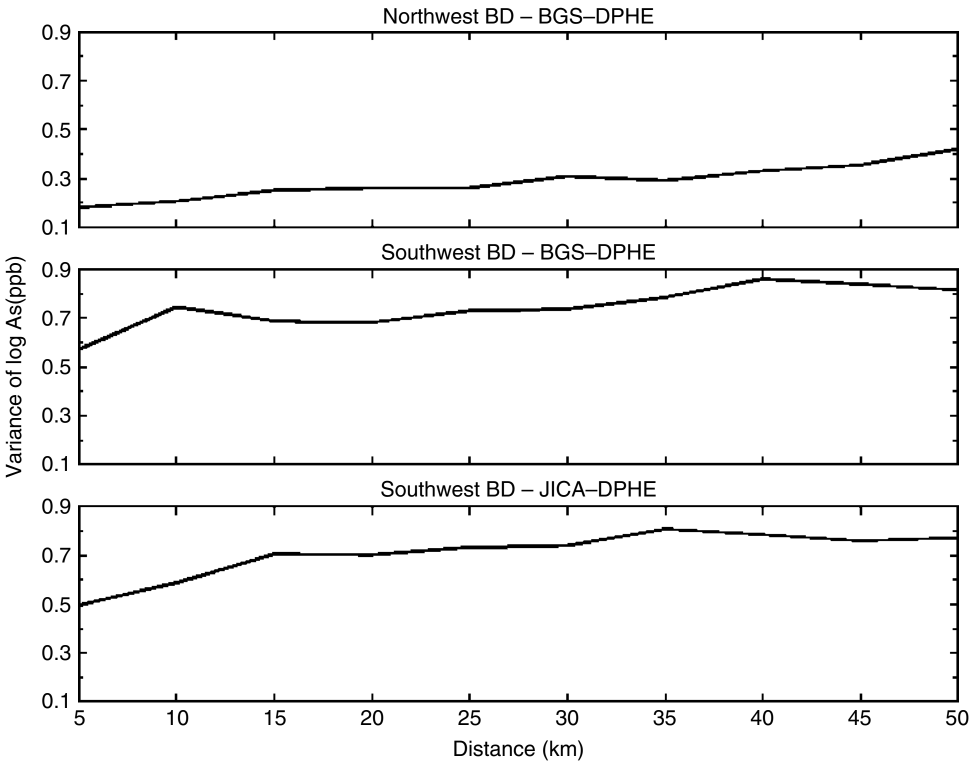

Figure 5. Isotropic variogram for northwest and southwest BD. Uppermost panel: variogram computed from the BGS–DPHE for northwest BD; middle panel: variogram computed from the BGS–DPHE for southwest BD; lowermost panel: variogram computed from the JICA–DPHE for southwest BD

Table II. Correlation lengths for spatial dependence of As in northwest and southwest BD

	Correlation length parameter *a* (km)		
	East–west	North–south	Isotropic
Northwest BD (BGS–DPHE)	158·80	78·21	87·21
Southwest BD (BGS–DPHE)	52·00	36·71	113·51
Southwest BD (JICA–DPHE)	36·01	23·91	83·61

emerged over the last 3 years since the BGS–DPHE survey in 1999. A higher correlation length signifies higher spatial dependence of As concentration. However, interpretation of the isotropic correlation lengths is conditioned upon the campaign chosen. For example, for northwest BD, because most wells are usually below the safe limit, the BGS–DPHE would seem to suggest that, if a shallow well is characterized as unsafe, the chance of locating a similar unsafe well in the vicinity is lower than that for southwest BD. The more recent JICA–DPHE campaign, however, rebuts this suggestion, as its correlation length for southwest BD is found to be similar to that of BGS–DPHE for northwest BD.

A different picture emerges on spatial dependence when anisotropy is considered. Figure 6 presents the anisotropic variogram for northwest BD and southwest BD. Sharp contrasts between the north–south variograms and the east–west variograms are observed. The variability, in general, appears considerably subdued in the east–west direction compared with the north–south direction. This is explained as follows.

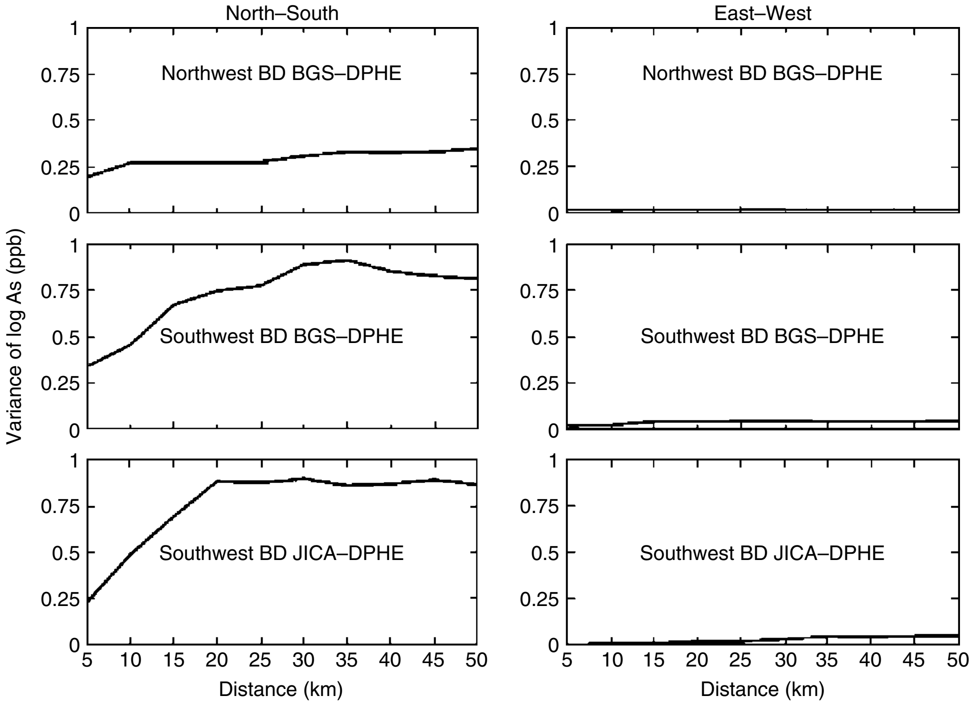

Figure 6. Anisotropic variogram for northwest and southwest BD. Left panels: north–south direction; right panels: east–west directions

Referring to Figure 4 for the district-mean As concentrations, we observe that the highest As concentrations are typically observed in the east–west direction for both northwest and southwest BD, with a minimum of variability and a high mean. For the north–south direction, the converse is observed. In terms of correlation lengths in northwest BD, the east–west length is double (158·80 km) that of the north–south length (78·21 km), whereas for the southwest the ratio of correlation lengths (east–west to north–south) ranges from 1·42 (BGS–DPHE) to 1·51 (JICA–DPHE). Again, owing to the more up-to-date measurement protocols, we assume the anisotropic character depicted by the JICA–DPHE survey to be a more refined representation of southwest BD than what was previously known from the BGS–DPHE survey. Anisotropy seems to suggest that the 'plume' of regionalized As contamination is stretched more in the east–west direction than the north–south direction. However, the most useful pattern deciphered from this anisotropy in the context of remediation is for northwest BD. There appears to exist approximately twice as much probability of locating an unsafe well near an identified unsafe well in the east–west direction than in the north–south direction. Hence, if contaminated shallow wells in northwest BD were to be replaced by safe deep wells, the network for planned drilling around the contaminated regions should take this anisotropy into account. On the other hand, for southwest BD, since the shallow aquifers are already contaminated with high As concentrations, the anisotropic property may not be as useful. However, if one were to take advantage of the fact that the depth where the highest contamination of wells is usually observed is around 40–50 m, then one should preferably try locating existing and potentially safe shallow wells deeper than 50 m in the north–south direction than the east–west direction. We may also postulate a similar rule of thumb for the existence of safe wells. That is, if a new safe well is needed to be drilled near the location of a previously identified safe well, then the east–west direction would be the more logical direction in which to proceed. It is appropriate to caution, however, that the rules of thumb postulated herein may not be representative at the sub-regional scale, as the anisotropic variography analyses did not pertain to measurement scales smaller than 5 km. Hence, a high As small town like Chapai Nawabganj (of 5 km × 5 km approximate dimensions)

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and its surrounding suburban areas may not derive much utility from these rules of thumb for designing a municipal water supply strategy.

CONCLUSIONS

A statistical characterization of the As contamination scenario in shallow tube wells (depth <150 m) of western Bangladesh is presented. Comparisons are made in light of geological differences (Pleistocene versus Holocene deposits/northwest versus southwest) and As measurement protocols (FK versus AAS). We characterized the error structure of the semi-quantitative FK As measurements using collocated AAS As measurements as reference from a set of 307 wells located in southwest Bangladesh. The probability of the FK method for successful detection of a well sample as unsafe (safe) was found to be 96·9% (12·5%) and 95·2% (80·3%) for the WHO and Bangladesh safe limits, respectively. Similarly, the probabilities of false alarms and false hopes for WHO (Bangladesh) safe limits were found to be 3·1% (4·8%) and 87·5% (19·7%), respectively. The depth where the highest fraction of wells exceeded a given safe limit could still be inferred accurately by FK measurements. A simple bias adjustment procedure on FK As data did not result in a more accurate characterization of depth distribution of As. This probably indicates that the database comprising 1·3 million FK As measurements that is currently available may indeed be sufficient to characterize the probable depth of highest As contamination for local regions, even though the modal As contamination rate (i.e. highest fraction of contaminated wells) may exhibit an error of about 50%. This partial utility of the FK method may, therefore, allow villagers and local water authorities to be aware of the risk of contamination for a shallow well provided that its depth is accurately known by the owner. Furthermore, a twofold strategy of initial screening of wells by the FK method (as a fast-running and inexpensive proxy) followed by the more reliable but costly AAS testing of FK-screened wells can potentially reduce operating costs during remediation efforts at the regional level.

Anisotropy in the spatial dependence for northwest BD (comprising mainly Pleistocene deposits) was found to be stronger than southwest BD (comprising mainly Holocene deposits). The correlation length for As concentration in the east–west direction of northwest Bangladesh was found to be almost twice (158·80 km) that of the north–south direction (78·21 km). For the southwest region, the ratio of east–west to north–south correlation lengths ranged from 1·40 to 1·50. Because northwest BD is already known to have the lowest concentrations of As countrywide, knowledge of this anisotropy supports the wisdom of drilling twice as more deep wells in the proximity of an unsafe shallow well in the north–south direction than in the east–west direction. We may hypothesize a similar rule of thumb for safe wells; i.e. if a new safe well is needed to be drilled near the location of a previously identified safe well, then the east–west direction would be the more logical direction in which to drill. Such rules of thumb, however, may not be useful at the sub-regional scale in high As pocket-like regions, such as the town of Chapai Nawabganj. Findings from this study are potentially useful in setting priority areas for emergency testing, distributing remediation resources equitably and formulating a regional water resources strategy for western Bangladesh.

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REFERENCES

Abernathy CO, Calderon RL, Chappel WR. 1997. *Arsenic Exposure and Health Effects*. Chapman and Hall: London.

Ahmed MF. 2003. *Arsenic Contamination: Bangladesh Perspective*. ITN/BUET: Dhaka.

Ahmed MF. 2002. *Arsenic Mitigation in Bangladesh. International Workshop in Bangladesh, Dhaka, 14–16 January*. ITN/Centre for Water Supply and Waste Management, BUET: Dhaka.

Alam MK, Hassan A, Khan M, Whitney JW. 1990. *Geological Map of Bangladesh*. Geological Survey of Bangladesh.

Bhattacharya P, Jacks G, Ahmed KM, Routh J, Khan AA. 2002. Arsenic in groundwater of the Bengal delta plain aquifers in Bangladesh. *Bulletin of Environmental Contaminant Toxicology* **60**: 538–545.

Biswas BK, Dhar RK, Samantha G, Mandal BK, Chakraborti D, Faruk I, Islam KS, Chowdury M, Islam A, Roy S. 1998. Detailed study report of Samta, one of the arsenic-affected villages of Jessore District, Bangladesh. *Current Science* **74**: 134–145.

BRAC. 2000. *Combating a deadly menace: early experience with a community-based arsenic mitigation project in Bangladesh*. Research Monograph Series 16, Research and Evaluation Division, Dhaka, Bangladesh.

Burgess WG, Burren M, Perrin J, Ahmed KM. 2000. 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*, Hiscock KM, Rivett MO, Davison RM (eds). Geological Society of London Special Publication 193. Geological Society Publishing House: Bath; 145–163.

DPHE-UNICEF. 2001. *Monthly progress report*, December, Dhaka, Bangladesh.

JICA–DPHE. 2002. *The study on the groundwater development of deep aquifers for safe drinking water supply to arsenic affected areas in western Bangladesh, final report, (Book 1 & 2)*. Department of Public Health Engineering, Government of Bangladesh, Dhaka, Kokusai Kogyo Co. Ltd & Mitsui Mineral Development Engineering Co., Ltd.

Harvey CF, Swartz CH, Badruzzaman ABM, Keon-Blute N, Yu W, Ali MA, Jay J, Beckie R, Niedan V, Brabander D, Oates PM, Ashfaque KN, Islam S, Hemond HF, Ahmed MF. 2002. Arsenic mobility and groundwater extraction in Bangladesh. *Science* **298**: 1602–1606.

Hossain F, Anagnostou EN. 2004. Assessment of current passive microwave and infrared-based satellite rainfall remote sensing for flood prediction. *Journal of Geophysical Research* **109**(D07102): DOI: 10·1029/2003JD003986.

Kinnburgh DG, Smedley PL (eds). 2001. *Arsenic contamination of groundwater in Bangladesh*, vols 1–4. British Geological Survey Report WC/00/19. British Geological Survey: Keyworth. http://www.bgs.ac.uk/arsenic/Bangladesh [10 January 2004].

McArthur JM, Ravenscroft P, Safiullah S, Thirlwall MF. 2001. Arsenic in groundwater: testing pollution mechanisms for sedimentary aquifers in Bangladesh. *Water Resources Research* **37**(1): 109–117.

Meharg AA, Rahman MM. 2003. Arsenic contamination of Bangladesh paddy fields. *Environmental Science Technology* **37**: 229–234.

Mukherjee AB, Bhattacharya P. 2002. Arsenic in groundwater in the Bengal delta plain: slow poisoning in Bangladesh. *Environmental Reviews* **9**: 189–220.

Nickson RT, McArthur JM, Burgess W, Ahmed KM, Ravenscroft P, Rahman M. 1998. Arsenic poisoning of Bangladesh groundwater. *Nature* **395**: 338.

Rahman MM, Mukherjee D, Sengupta MN, Chowdury UK, Lodh D, Chanda CN, Roy S, Selim M, Quamruzzaman Q, Milton AH, Shadullah SM, Rahman MT, Chakraborti D. 2002. Effectiveness and reliability of arsenic field testing kits: are the million dollar screening projects effective or not? *Environmental Science and Technology* **36**: 5385–5394.

Tanabe K, Akiyoshi Y, Hironaka H, and Yokota H. 1997. Arsenic contamination around Ganges River and the applicability of a field kit by Hironaka to quantify arsenic. In *Abstracts in the Meeting of Kyushu and Chugoku, Shikoku Branches of the Chemical Society of Japan*, no. 102.

van Geen A, Zheng Y, Vesteeg R, Stute M, Horneman A, Dhar R, Steckler M, Gelman A, Ahsan H, Graziano JH, Hussain I, Ahmed KM. 2003. Spatial variability of arsenic in 6000 tube wells in a 25 km^2 area of Bangladesh. *Water Resources Research* **39**(5): 1140. DOI: 10·1029/2002/WR001617.

Yu WH, Harvey CM, Harvey CF. 2003. Arsenic groundwater in Bangladesh: a geostatistical and epidemiological framework for evaluating health effects and potential remedies. *Water Resources Research* **39**(6): 1146. DOI: 10·1029/2002WR001327.

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