Application of geostatistics with Indicator Kriging for analyzing spatial variability of groundwater arsenic concentrations in Southwest Bangladesh

(2011) *Journal of Environmental Science and Health, Part A: Toxic/Hazardous Substances & Environmental Engineering* 46, 11, 1185-96

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This article seeks to explore the spatial variability of groundwater arsenic (As) concentrations in Southwestern Bangladesh. Facts about spatial pattern of As are important to understand the complex processes of As concentrations and its spatial predictions in the unsampled areas of the study site. The relevant As data for this study were collected from Southwest Bangladesh and were analyzed with Flow Injection Hydride Generation Atomic Absorption Spectrometry (FI-HG-AAS). A geostatistical analysis with Indicator Kriging (IK) was employed to investigate the regionalized variation of As concentration. The IK prediction map shows a highly uneven spatial pattern of arsenic concentrations. The safe zones are mainly concentrated in the north, central and south part of the study area in a scattered manner, while the contamination zones are found to be concentrated in the west and northeast parts of the study area. The southwest part of the study area is contaminated with a highly irregular pattern. A Generalized Linear Model (GLM) was also used to investigate the relationship between As concentrations and aquifer depths. A negligible negative correlation between aquifer depth and arsenic concentrations was found in the study area. The fitted value with 95 % confidence interval shows a decreasing tendency of arsenic concentrations with the increase of aquifer depth. The adjusted mean smoothed lowess curve with a bandwidth of 0.8 shows an increasing trend of arsenic concentration up to a depth of 75 m, with some erratic fluctuations and regional variations at the depth between 30 m and 60 m. The borehole lithology was considered to analyze and map the pattern of As variability with aquifer depths. The study has performed an investigation of spatial pattern and variation of As concentrations.

Keywords: Arsenic, geostatistics, GLM, Indicator Kriging, Bangladesh.

Introduction

Groundwater in Bangladesh is reportedly found to be contaminated with toxic levels of arsenic (As) that threaten the health of millions of people in Bangladesh. The impact of As poisoning on human health in Bangladesh has been alleged to be the "worst mass poisoning in human history".[1] Since the discovery ofAs in groundwater in 1993 by the Department of Public Health Engineering (DPHE), As contamination has been increasing at an alarming rate, and the risk is spreading all over the country. The extensive use of As-contaminated groundwater for drinking and cooking threatens the health of about 70 million people in 61 out of 64 districts in Bangladesh.[2] Arsenic contaminated wells across the country are thought to be a hazard to human health. One estimate is that millions of people may die or suffer from the very serious consequences of consuming As.[3]

There is a complex pattern of spatial variability of As concentrations in groundwater, with significant differences between neighboring wells, trends at the regional scale, and also

changes with depth underground.[4,5–10] Yu et al.[3] find a small statistically insignificant positive correlation between observed As concentrations and shallow tubewell density. Mukherjee et al.[11] observed elevated dissolved As (>10 μg/L) in a majority of the deep groundwater samples in West Bengal, India and the maximum concentration was recorded at 137 μg/L. In addition, very little is known about changes in As concentration over time,[3] but a temporal variation of groundwater As concentrations is thought to be significant.[12–13]

The Indicator Kriging (IK) proposed by Journel[14] is one of the most efficient nonparametric methods in geostatistics.[15] IK is a spatial interpolation technique devised for estimating a conditional cumulative distribution function at an unsampled location.[16] It has become the basis of some estimate algorithms and sequential indicator simulations. Meliker et al.[17] investigated the validity of different spatial models. IK can model for estimating the probability distribution of spatial variables on the basis of surrounding observations. In this study, an IK approach was adopted for analyzing the spatial pattern of As concentrations in Southwest Bangladesh.

Because of the distribution of groundwater As is extremely heterogeneous in both vertical and horizontal dimensions, a proper policy formulation to extract groundwater with new tubewells is problematic. Mapping the geographical pattern of groundwater As concentrations with a spatial dimension is the focal issue of this study, and emphasis is laid on the relationships between As concentrations and aquifer depths as well as tubewell age. Borehole lithology, the new data for this paper, is considered to analyze the pattern of As concentrations at different stratigraphy.

Earlier we reported for the same study site analyses of spatial risk fromAs poisoning andmainly focused ourwork on the spatial risk pattern of groundwater As poisoning analyzed with Ordinary Kriging (OK) method.[18] However, in the present study described in this paper the subsurface geology of the study site has been considered with the inclusion of a number of spatial analytical figures. Geostatistics with IK interpolation technique and Generalized Linear Model (GLM) which are the main data analysis methods were employed in order to investigate the pattern of As concentrations and its spatial variability. This spatial variation of As concentrations will be helpful in formulating spatial policy to mitigate As poisoning in highly contaminated areas and so reducing the "health risk".

Materials and methods

Study area and geology

The data for this study were collected from Ghona *Union* (the fourth-order local government administrative unit in Bangladesh) of Satkhira District in the southwest of Bangladesh near to the border with India (Fig. 1). The study area comprises nine administrative wards covering an area of 17.26 km2 and had a population of 13,287 in 1991.[19] The study area is geologically a part of the quaternary deltaic sediments of the Ganges alluvial and tidal plains.[20] The surface geology of the study area mainly comprises (a) the Ganges alluvial plain, which covers the middle part of the study area (about one-third); and (b) the northern and southern parts (two-thirds) that lies in the Ganges tidal plains. The British *Khal* (canal) and theMahmudpur *Khal* are the two main rivers flowing through the study area. The study area has been dominated by irrigated agriculture for the last few decades.

The subsurface geology of the study area has complex inter-fingerings of coarse and fine-grained sediments from numerous regressions and transgressions throughout geologic

time.[21] The Holocene sedimentary succession, in the study area, shows a fining upward sequence from medium to fine sand, silt and finally to clay. The aquifer of this region is mostly unconfined to leaky confined and groundwater occurs within a few meters of the surface. Two distinct aquifers occur all over the study area. The deep aquifer in the study area is at depths of ≥144.5 meters and the shallow aquifer at depths of <144.5m (Fig. 1). The aquitard separating the shallow aquifer from the deep aquifer is a continuous one and increases in thickness towards the south and at places the thickness is about 100 m.[22] The upper part of the shallow aquifer is comprised of fine to very fine sands and the lower part is medium to course sand. Arsenic is generally found in shallow aquifer; while the deeper aquifer has not so far been found to be severely contaminated, other than in a few cases.

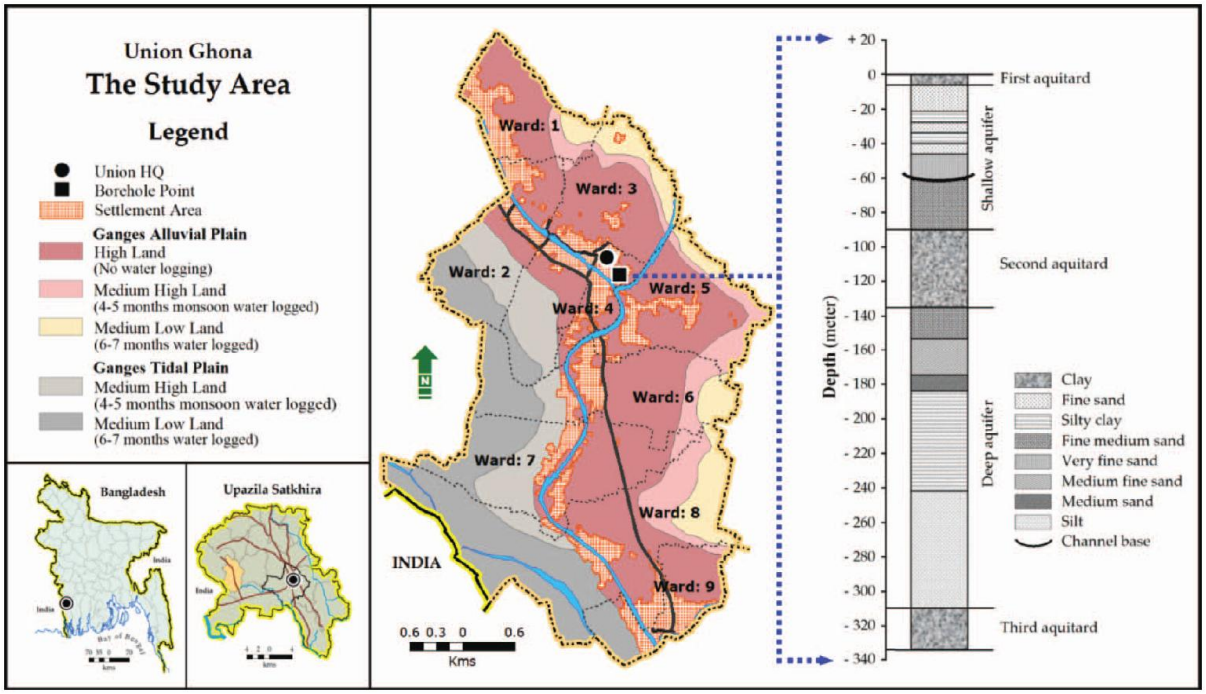

Fig. 1. The study area with lithology of a borehole: Ghona *Union* at Southwest Bangladesh. The borehole is located near the Ghona *Union* Headquarters (color figure available online).

Arsenic and attribute data

In most quantitative inquiry, the dominant sampling strategy is probability sampling. Tubewell screening is important priority work for As data collection. Which tubewells would be screened and how many? This was an important and sensitive issue in the context of present arsenic situation in Bangladesh. Our previous experiences in this regard were taken into account, and all the tubewells were screened. Moreover, since groundwater As concentrations are uneven in space and time dimension, all the 375 tubewells in the study area were analyzed. The geographical location of each tubewell in the study area was plotted on *mouza* maps (lowest level administrative territorial unit in Bangladesh) having a scale of 1:3960. Field test kits (FTK) are easy to use in analyzing groundwater As concentrations and are cost-effective, but their results are less reliable and less accurate than laboratory methods.[13,18,23–25] In addition, FTK results are not accurate enough to permit testing at theWHOpermissible limit and sometimes even the Bangladesh DrinkingWater Standard (BDWS).

For evaluation of the reliability and accuracy of the As data, all the collected water samples (n = 375) were analyzed with FI-HG-AAS method from the School of Environmental Studies

of Jadavpur University, Kolkata, India. To prevent adsorption losses, the collected samples were preserved by acidification with a 0.05 mL of concentrated nitric acid (14 M) in each 10mLofwater sample and placed in a refrigerator at a temperature below 4◦C prior to analysis. The method is characterized by high efficiency, low sample volume, reagent consumption, improved tolerance of interference, and rapid determination.[26–27]With a 95 % confidence interval, the minimum detection limit of the FI-HG-AAS method is 0.001mg/L, and the quantification limit is 0.003 mg/L,[28] which is excellent for As research. Along with the As content in water, two main attributes were collected for each tubewell: (a) tubewell depth and (b) tubewell installation year. All of the tubewell owners (375 tubewells) were asked for information about these attributes of their tubewells through a questionnaire survey. This attribute information was stored as records (rows) in a relational database. The map features (e.g., point, line, and polygon) were used for geoststistical analysis and GIS (Geographical Information Systems) mapping. The incorporated borehole litholog data was collected from the Department of Public Health Engineering (DPHE) office in Satkhira in 2009. The litholog data was then compiled and incorporated with arsenic concentrations, tubewell depths, aquifer depths, and age of each tubewell.

Geostatistics and indicator kriging

The geostatistical approach is a distribution-free procedure and is based on a theory of regionalized variables whose values vary from place to place.[29–30] It relies on both statistical and mathematical methods to create surfaces and to assess the uncertainty of predictions. Geostatistics represents an appropriate method of prediction[15,31] and is widely used for spatial estimation taking spatial variability into account.

Variogram provides a means of evaluation of the attributes in which each estimate is a weighted average of the observed values in the neighborhood. The weights mainly depend on fitting the variogram to the measured points. The variogram quantifies the spatial variability of the random variables between two sites. The experimental variogram is fitted with a theoretical model, $\gamma(h)$, which may be spherical, exponential or Gaussian, to determine the nugget effect ($C0$), the sill ($C0 + C1$) and the range (a). The variogram can be computed in different directions to detect any spatial anisotropy of the spatial variability.[32] This study adopted a geometric anisotropic model, which yields variograms with the same structural shape and variability (sill+nugget) but a direction-dependent range for the spatial correlation.[33] The general equation for estimating prediction value, $\hat{Z}(S_0)$, is given by

$$\hat{Z}(S_0) = \sum_{i=1}^{N} \lambda_i Z(S_i) \qquad (1)$$

Where, $\hat{Z}(S_0)$ = prediction value for location, S_0;

N = number of measured sample points surrounding the prediction location;

λ_i = the weight obtained from fitted variogram; and

$Z(S_i)$ = observed value at location S_i.

A kriging treatment quantifies the variability of As in the form of a semivariogram, which graphically expresses the relationship between the semivariance and the sampling distance.[34] The semivariogram, $\hat{}\,\gamma(h)$, is half the average squared difference between pairs of data $Z(xi)$ and $Z(xi + h)$ at locations xi and $xi + h$. An estimate of the semivariogram with $N(h)$ the number of sampling pairs separated by a distance of h(lag) is given by the following equation:

$$\hat{\gamma}(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} \left\{ Z(x_i) - Z(x_i + h) \right\}^2 \qquad (2)$$

IK is an advanced nonparametric geostatistical method due to its ability to take data uncertainty into account. IK makes no assumption regarding the distributions of variables, and a 0–1 indicator transformation of data is adopted to ensure that the predictor is robust to outliers.[35] In an unsampled location, the values estimated by IK represent the probability that does not exceed a particular threshold. Therefore, the expected value derived from indicator data is equivalent to the cumulative distribution function of the variable.[36] IK provides an estimate of the cumulative distribution of the data set by calculating conditional probabilities. These probabilities can be estimated by transforming the variables to a one or zero, depending upon whether they fall above or below a cutoff level:[37]

$$i(X;Z) = \begin{cases} 1 \, if \;\; Z(X) \le Z_k \\ 0 \, if \;\; Z(X) > Z_k \end{cases} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

Where, Z_k is the cutoff level.

By using kriging, the interpolated indicator variable at any point *X*0 can be estimated by:[38]

$$I_k^*(X_0) = \sum_{j=1}^{n} \gamma_{j0} i_k(X_j) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

Where, I_k^* = the estimate of the conditional probability at X_j;

γ_{j0} = the kriging weight for the indicator value at point X_j.

The conditional probability in this case is defined as:

$$Prob\left[Z(X_0) \le Z_K / (Z_j; j = 1, \ldots\ldots, n) \right] \quad \ldots\ldots\ldots\ldots\ldots \quad (5)$$

By varying Z_k, the cumulative probability can then be constructed.

IK determines the probability of the As indicator in the study area by using the samples in the neighborhood. To conduct IK, As concentrations were transformed into an indicator variable and the variogram function was evaluated in horizontal directions to identify the anisotropic variation present in groundwater As concentrations. Some three threshold values (i.e., the first quartile, median and the third quartile)were selected forAs indicator analysis (Table 1). An omnidirectional variogram was used to analyze the spatial structures of As concentration. A lag increment of 1/2 km was adopted to obtain a stable variogram structure. The spatial As probability map was analyzed and interpolated in ArcGIS (version 9.2).

Table 1. Fitted parameters of the spherical model for three threshold values of the arsenic indicator variable.

Threshold	*Nugget effect* (C_o)	*Partial sill* (C_1)	*Sill* ($C_0 + C_1$)	*Range (a)* (km)	*Major range* (km)	*Minor range* (km)	*Anisotropic ratio*
First (0.1405 mg/L) (1st Quartile)	0.1658	0.0298	0.1956	2.437	3.482	1.279	2.722
Second (0.241 mg/L) (Median Value)	0.1439	0.113	0.2569	0.815	3.298	0.781	4.222
Third (0.3267 mg/L) (3rd Quartile)	0.1232	0.0773	0.2005	0.805	2.790	0.833	3.349

The IK prediction map was developed with spherical variogram fit. The experimental variogram was computed from the As data and a mathematical model was fitted to As values by weighted least-squares approximation. The spherical model was used to fit the raw semivariogram:[39]

$$\hat{\gamma}(h) = \begin{cases} 0 & h = 0 \\ C_0 + C_1\left[\frac{3}{2}\left(\frac{h}{a}\right) - \frac{1}{2}\left(\frac{h}{a}\right)^3\right] & 0 < h < a \\ C_0 + C_1 & h \geq a \end{cases} \quad \ldots\ldots\ldots\ldots \quad (6)$$

where, *C0* is the nugget variance, and the lag, *h* required to reach the sill (*C0* + *C*1) is called a range, *a*. *Nugget* is a measure of spatial discontinuity at small distances, *sill* is an estimate of sample variances under assumption of spatial independence, and *range* is the distance at which sample data is spatially independent. In producing prediction maps for spatial As concentrations with IK prediction method, different properties for cross-validation in terms of semivariogram and search neighbourhood were used in the interpolation (Fig. 2).

Generalized linear models

The GLM was used to identify the association between As concentrations and aquifer depths as well as tubewell installation age. The GLM is a mathematical extension of linear models that do not force data into unnatural scales, and thereby allow for non-linearity and non-constant variance structures in the data.[40–43] They are based on an assumed relationship (link function) between the mean of the response variable and the linear combination of the explanatory variables. Hypothesis testing applied to the GLM does not require normality of the response variable, nor does it require homogeneity of variances. Since As data are not distributed normally, the GLM was used for this paper. The maximum likelihood estimation technique is an important advent in the development of GLM.[42] TheNewton-Raphson (maximumlikelihood) optimization technique was used in this paper to estimate the GLM and the STATA statistical software was utilized to calculate the GLM.

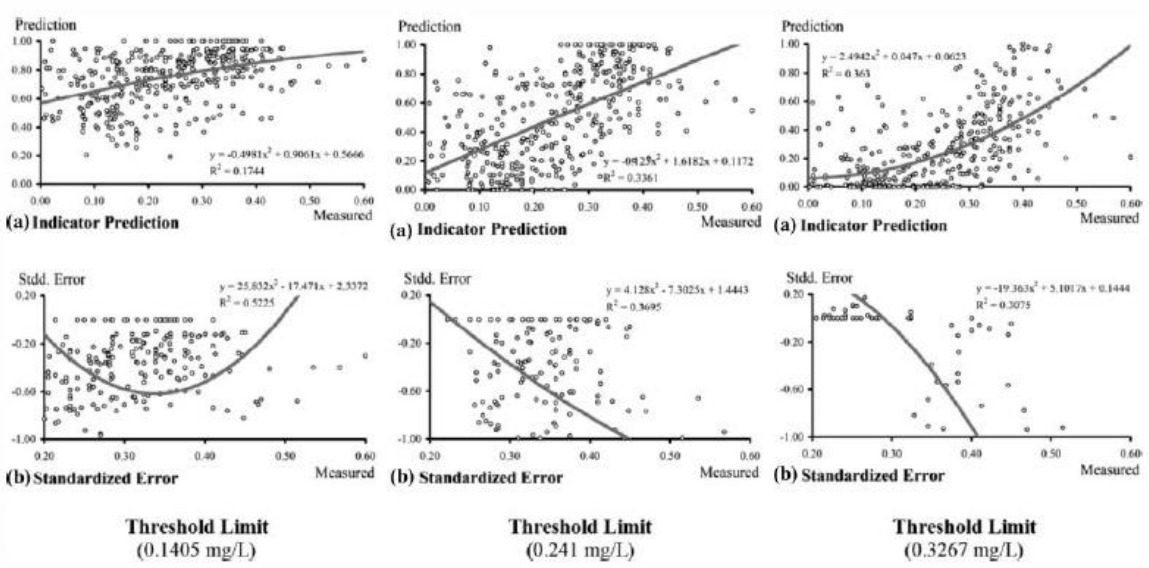

Fig. 2. Properties of cross-validation and prediction-error for IK prediction method for spatial arsenic analysis.

Performance assessment

In the prediction maps of arsenic concentrations, cross validation was used to compare the prediction performance of the IKinterpolation algorithm (Fig. 2). Cross-validation indicators and the model parameters (nugget, sill, and range) help us to choose a suitablemodel for the prediction maps of arsenic concentrations. In the procedure, arsenic concentration is estimated successively for each sampled tubewell using the known neighbours. The cross-validation was applied to improve and to control the quality of the applied geostatistical model and thus the results of the spatial analysis. The differences between measured and predicted values provide a quality control for the model of computation for arsenic concentrations. The difference between measured and estimated values was obtained by cross validation. Based on the cross-validation results of the data exploration and the variogram analysis, the measured data are converted to surface maps. IK is used to divide values at locations to three different categories by means of a threshold limits. The observed values $Z(U)$ are then compared with the interpolated ones $Z^*(U)$ using the performance measures the bias:[44]

$$Bias = \frac{1}{n}\sum_{i=1}^{n}[Z^*(U_i) - Z(U_i)] \qquad (7)$$

And the root mean square error normalized with the observed average:[44]

$$RMSE = \frac{1}{\bar{Z}}\sqrt{\frac{1}{n}\sum_{i=1}^{n}[Z^*(U_i) - Z(U_i)]^2} \qquad (8)$$

Interpolation usually leads to a smoothing of the observations and thus to a loss of variance. In analyzing the data with IK prediction method, we found the average standard error of arsenic data to be 0.3903 with root mean square of 0.3964. The figures for mean standardized error were analyzed by −0.0034 and 1.023 for root mean square standardized. Figure 2 shows the details about the prediction error of arsenic concentrations with the IK prediction method.

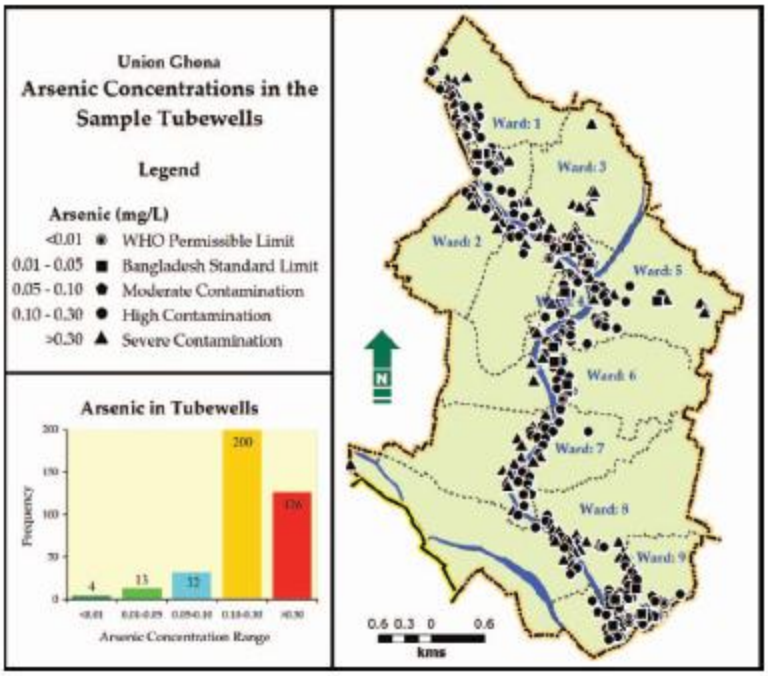

Fig. 3. The pattern of arsenic concentrations in all the tubewells in the study area. Some 375 tubewells were investigated for arsenic concentrations (color figure available online).

Results and discussion

Scale of arsenic concentrations

There has been a heterogeneous distribution of groundwater As concentration in the study area (Fig. 3). The term "contamination" in this article refers to the elevated concentrations of As above the BDWS. Arsenic concentrations in the study area range between 0.003 mg/L and 0.600 mg/L with a mean concentration of 0.238 mg/L and the standard deviation of 0.117 mg/L. [18] The study shows that only 4.50 % of the sample tubewells (17 out of 375) belong to the safe level and 95.5 % (358 out of 375) are said to be contaminated with As ≥0.05 mg/L (Table 2).

In the safe band, only four tubewells meet the WHO permissible limit (<0.01 mg/L) and 13 tubewells qualify at the BDWS (Table 2). In addition, in the contamination category, As concentrations range between 0.057 mg/L and 0.600 mg/L with a mean concentration of 0.248 mg/L and standard deviation (δn) of 0.109 mg/L (Table 2). It is noteworthy that the mean As concentration in this contamination category is 5 times higher than the BDWS and 25 times higher than the WHO permissible limit. [18] The pattern of concentrations varies considerably and unpredictably over distances of a few meters, notably about 46 % of tubewells are located within 25 m of each other within the settlement area of the study site.[18]

The measured As concentrations are 0.1405 mg/L, 0.241 mg/L and 0.3267 mg/L at the first, second (median) and the third quartile of the frequency distribution of As concentrations (Table 1). The omnidirectional variogram was adopted to analyze the geometric anisotropic variability. The anisotropic ratios (maximum range/minimum

Table 2. Pattern of groundwater arsenic concentrations with detailed classification in different aquifer levels and statistics.

Major groups	*Arsenic (mg/L)*	*Detailed classification*	*Shallow aquifer (<144.5 m)*	*Deep aquifer (≥144.5 m)*	*Total tubewells*
Safe level	<0.01	WHO Limit	–	4 (1.064)	04 (1.064)
	0.01–0.05	Bangladesh Standard Limit	7 (1.867)	6 (1.596)	13 (3.458)
Contamination level	0.05–0.1	Moderate contamination	30 (7.98)	2 (0.532)	32 (8.512)
	0.1–0.3	High contamination	200 (53.20)	–	200 (53.20)
	>0.3	Severe contamination	126 (33.516)	–	126 (33.516)
		Total	363 (96.558 %)	12 (3.192 %)	375 (100 %)
			$\bar{x} = 0.245$mg/L	$\bar{x} = 0.025$mg/L	$\bar{x} = 0.238$mg/L
			$\delta_n = 0.112$ mg/L	$\delta_n = 0.026$ mg/L	$\delta_n = 0.117$ mg/L
			$r = +0.126$mg/L	$r = -0.0769$mg/L	$r = -0.216$mg/LA

Data Source: Field survey, 2001.
(Figures in parentheses indicate the net percent of the sample tubewells).

range) range from 2.722 to 4.222 (Table 1). The variogram for IK with nugget effect, range and sill shows the local erratic variation of As concentrations. The minor range of continuity in the variogram with large nugget effect results from the high and uneven spatial continuity of As concentrations (Fig. 4). The upward trend between nugget and partial sill indicates some regional spatial trends in As variability and it is probably due to the subsurface redox gradient.

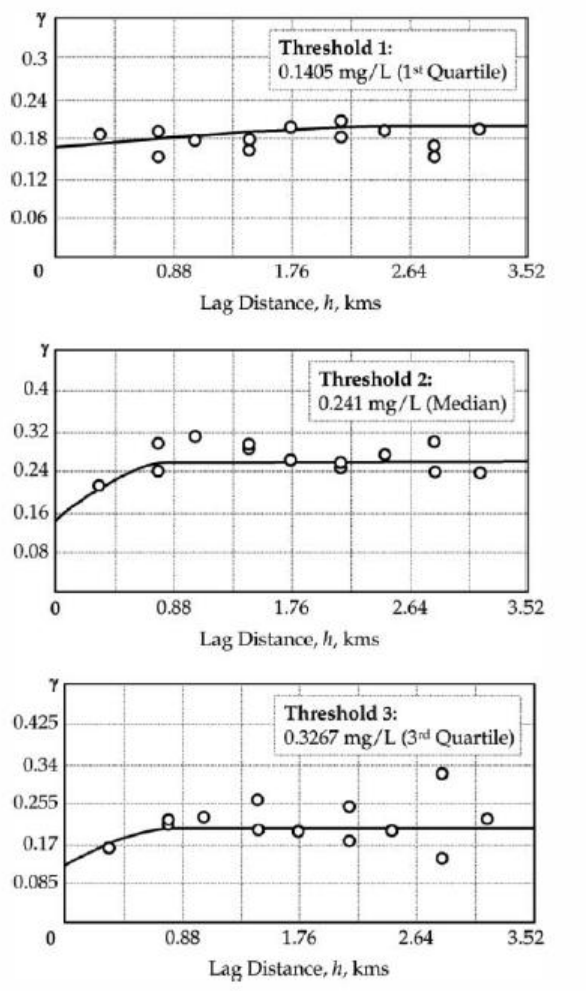

Fig. 4. Spherical variogram indicator and the fitting of theoretical models in horizontal directions for three threshold values (i.e., 0.1405 mg/L, 0.241 mg/L, and 0.3267 mg/L).

The probability map developed with the IK prediction method shows a highly uneven spatial pattern of As concentrations (Fig. 5). It is noted here that Figure 5 was prepared with IK prediction method. Three different threshold limits for As concentrations were used to develop the maps for Figure 5. Table 1 shows the details regarding this issue. Different threshold limits show different patterns of spatial As continuity in the study area (Fig. 5). There are visible discrepancies in spatial continuity of As concentrations in the central and eastern parts of the study area and the pattern is very uneven. The safe zones are mainly concentrated in the north, central and south part of the study area in a scattered manner. The central part of the study area is found to be low contaminated - in an area roughly corresponding to the Ganges alluvial floodplain. Contamination zones are found everywhere in the study area but with a decrease in the degree of contamination from west to east: they are largest in the low-lying area of the west and north-east. The south and southeast regions appear to show safe zones with some local variability. The west and northeast parts of the study area are mostly contaminated; while the southwest part of the study area is contaminated with a highly irregular pattern (Fig. 5). The safe zones are associated with the highest elevations of the area, and the contamination zones are on the west and northeast part where the elevation is low and agriculture predominant.

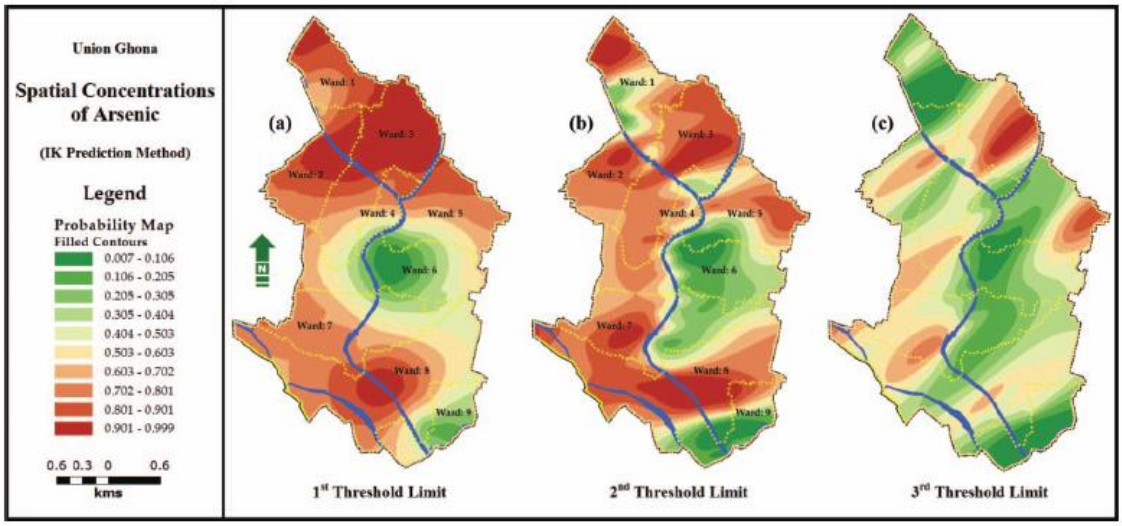

Fig. 5. Spatial pattern of arsenic concentrations prepared with IK prediction method: (a) arsenic concentration at threshold limit of 0.1405 mg/L; (b) at the threshold limit of 0.241 mg/L; and (c) at the limit of 0.3267 mg/L (color figure available online).

Arsenic concentrations with depth

Arsenic concentrations in the study area are highly uneven with depth (Fig. 6a). Tubewell depths range between 18 m and 200 m. Drawing upon the shallow aquifer (≤144.5 m) and deep aquifer (>144.5 m), about 97 % (n = 363) and 3.0 % (n = 12) tubewells have been recognized respectively (Table 2). The aquifer below the second aquitard is generally defined as the deep aquifer; while aquifer located in between the first and second aquitards is known as the shallow aquifer. The second aquitard is located between 92.0 and 144.5 m depth in the study site (Fig. 1). Of those using the shallow aquifer, only 1.87 % cent of the tubewells (n = 7) were found to be safe and about 94.7 % (n = 356)were contaminated,with amean concentration of 0.254 mg/L (Table 2). Moreover, in the deep aquifer, only eight tubewells (out of 12) failed the WHO standard and two tubewells failed the BDWS (Table 2). A negligible negative correlation between aquifer depth and As concentrations was found in the study area. The product moment of correlation value (r = −0.216) shows that there is a slow

decreasing tendency of As concentrations with increase of aquifer depth. The fitted value with 95%confidence interval shows a decreasing tendency ofAs concentrations with the increase of aquifer depth, but As concentration levels are low in tubewells tapping the deep

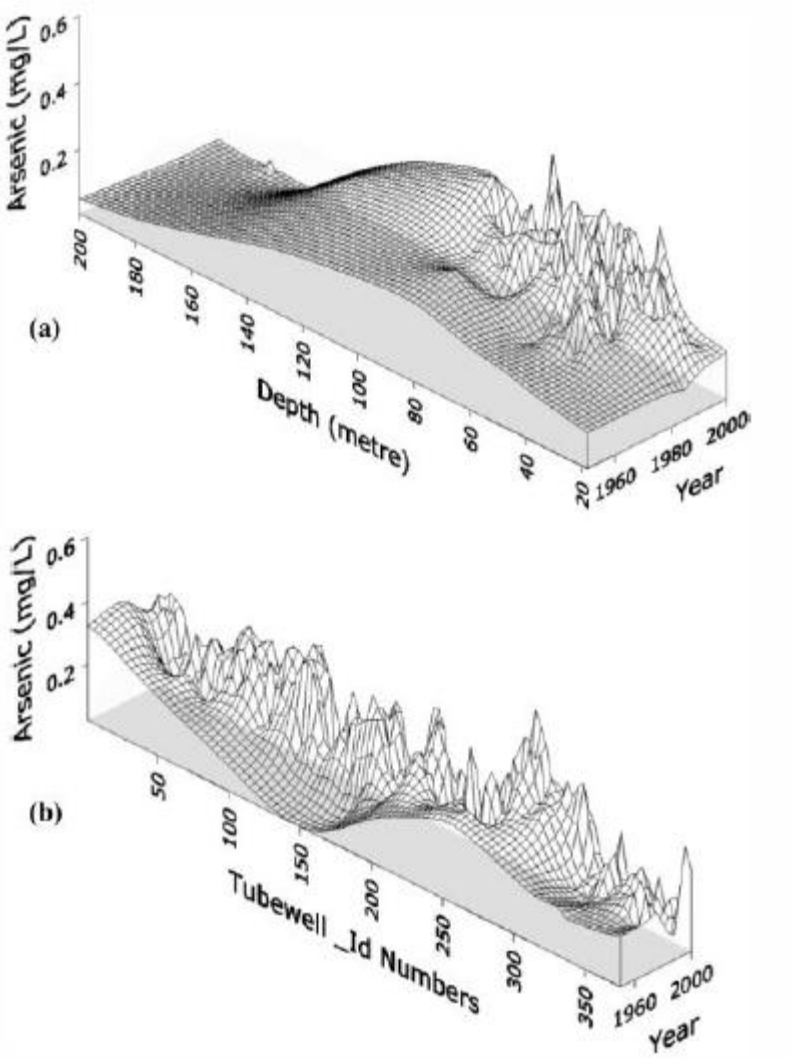

Fig. 6. Pattern of arsenic concentrations in the study area: (a) three dimensional view with arsenic-depth-time; and (b) arsenic concentration with tubewell installation year.

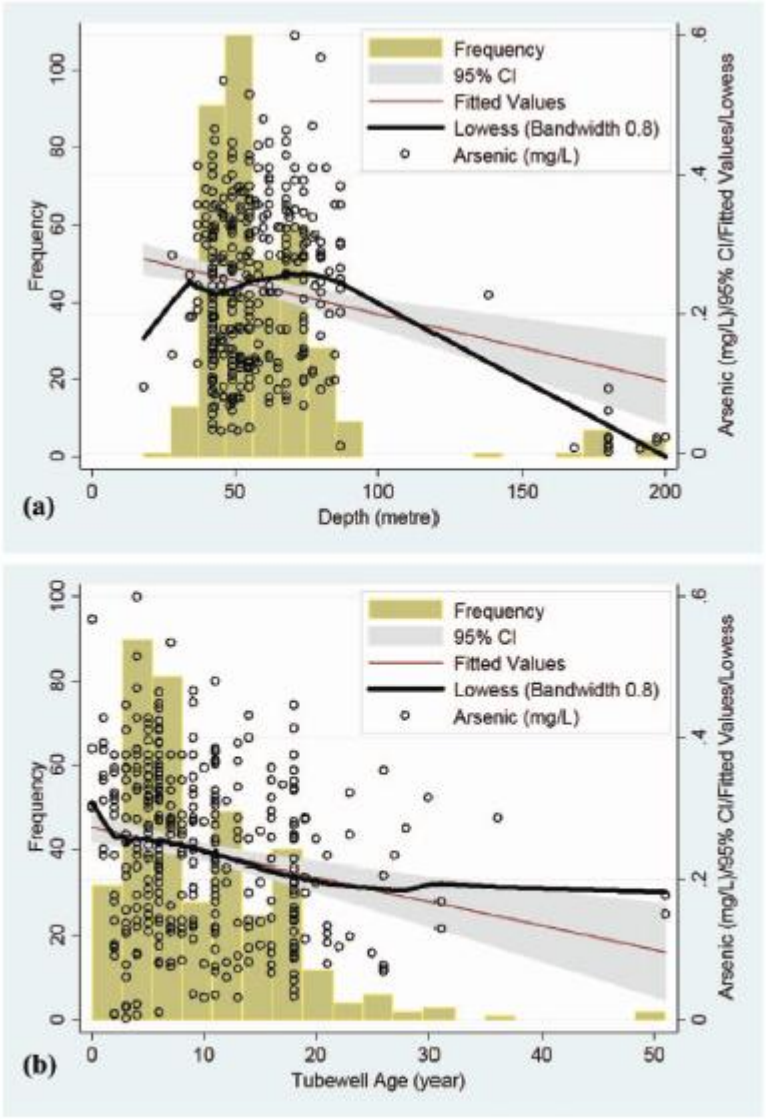

Fig. 7. Relationships between arsenic concentrations with aquifer depth and tubewell age: (a) concentrations in different aquifer depths; and (b) concentrations with tubewell age (color figure available online).

aquifer. In addition, the adjusted mean smoothed lowess curve with a bandwidth of 0.8 shows an increasing trend of As concentration up to a depth of 75 m, with some erratic fluctuations and regional variations at the depth between 30 and 60 m (Fig. 7a).

Marked regional variations and a considerable contrast in As concentrations are noticeable in the sub-surface geology (Table 3). Arsenic concentrations in the 'fine sand zone' (depth: 42.7–49.2 m) range from 0.032 mg/L at 45 m depth to 0.535 mg/L at 46mdepth, with amean concentration of 0.219 mg/L for 85 tubewells. In the 'very fine sandy zone' (depth: 49.3–65.6m), the value ranges between 0.032 mg/L at 51mdepth and 0.515 mg/L at 55mdepth with themean concentration of 0.243 mg/L for about two-fifths (n = 153) of the total sample tubewells. At the depth of 65.7–91.9 m (fine to medium sandy zone), As concentrations range between 0.011 mg/L at 85 m depth and 0.600 mg/L at 71m depth, having a mean concentration of 0.264 mg/L for 105 tubewells (Table 3).

It was found that the pattern of As concentration varies with grain-size distribution and that there is a lithological relationship with As concentrations. The '*r* ' values show very negligible positive associations between As concentrations at depths 42.7–49.2 m and 49.3–65.6 m; while a negligible negative association was found at depths of 65.7–91.9 m (Table 3).

Arsenic concentrations were also found to be highly uneven at certain depths (Fig. 8). At a depth of 42 m, a sharp variation in As concentrations was identified in 51 tubewells having a range between 0.034 and 0.428 mg/L, with a mean concentration of 0.2035 mg/L. At a depth of 55 m, a substantial variability in As concentrations for 28 tubewells was found: between 0.037 and 0.515 mg/L. At a depth of 74 m, a sharp variation was also identified in 25 tubewells having a range between 0.069 and 0.392 mg/L. Some nine tubewells have been found within a 135 m radius in the southern part of the study area that have values between 0.142 and 0.241 mg/L; while at a depth of 180 m, dissimilarity in As concentrations was found for seven tubewells having a range between 0.003 to 0.093 mg/L (Fig. 8).

Table 3. Arsenic concentrations in different borehole lithologic strata.

Borehole depth (m)	*Stratigraphy*	*Aquifer level*	*Tubewells*	*Safe (<0.05mg/L)*	*Contaminated (>0.05mg/L)*	*Concentration range*	*Mean ($\bar{x}$)*	*Std. deviation (δ_n)*	*Correlation (r-value)*
0–6.5	Clay	1st aquitard	–	–	–	–	–	–	–
6.6–22.9	Fine sand	Shallow aquifer	1	–	1	–	–	–	–
23.0–29.5	Silty clay	do	–	–	–	–	–	–	–
29.6–36.1	Fine sand	do	4	–	4	0.142–0.285	0.220	–	–
36.2–42.6	Silty clay	do	15	–	15	0.129–0.413	0.294	0.0790	–
42.7–49.2	Fine sand	do	85	3	82	0.032–0.535	0.219	0.1104	+0.1318
49.3–65.6	Very Fine sand	do	153	3	150	0.032–0.515	0.243	0.1150	+0.1724
65.7–91.9	Fine to Medium sand	do	105	1	104	0.011–0.600	0.264	0.1102	−0.0416
92.0–144.4	Clay	2nd aquitard	–	–	–	–	–	–	–
144.5–164.0	Fine to Medium sand	Deep aquifer	–	–	–	–	–	–	–
164.1–187.0	Fine to Medium sand	do	1	1	–	–	–	–	–
187.1–196.9	Medium sand	do	7	5	2	0.003–0.093	0.032	0.033	–
197.0–259.2	Silty clay	do	4	4	–	0.007–0.023	0.017	0.007	–
259.3–331.4	Silt	do	–	–	–	–	–	–	–
331.5–357.6	Clay	3rd aquitard	–	–	–	–	–	–	–
			375	17	358				

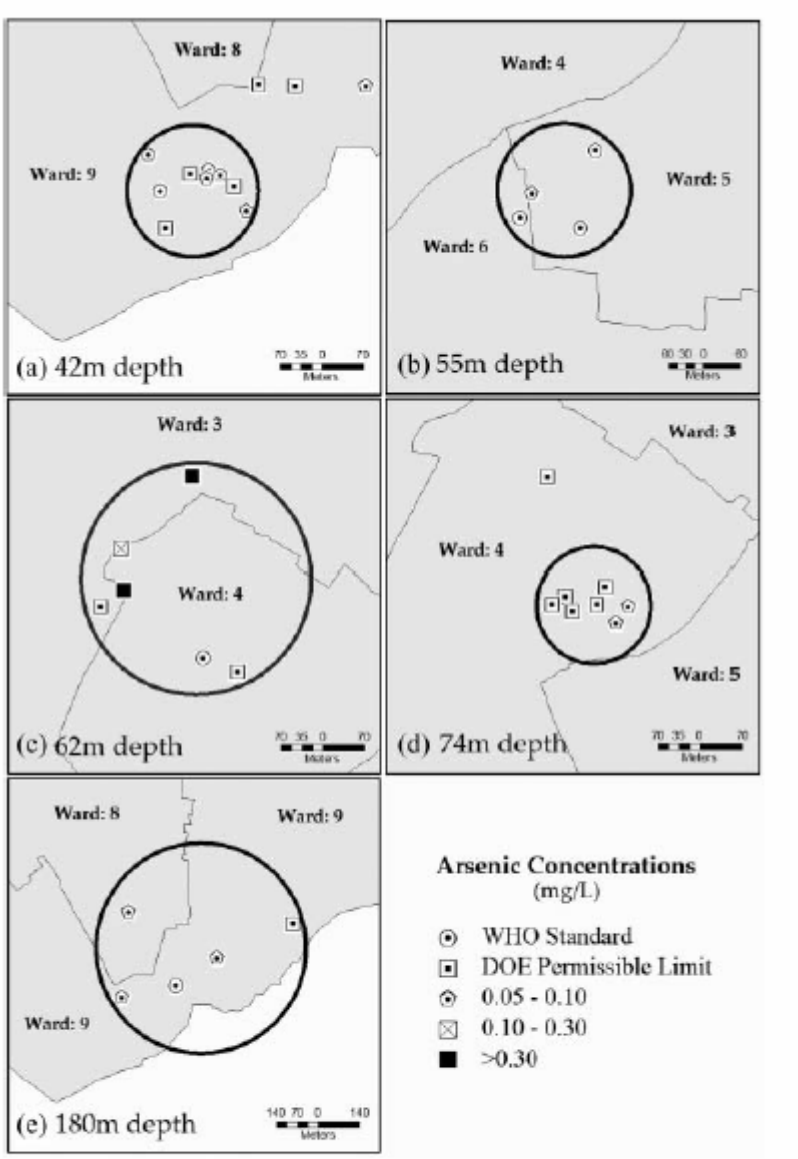

Fig. 8. The pattern of arsenic concentrations at certain depths in the study area. The symbology shows the qualitative variation of arsenic concentrations at very close distances within a certain depth in the aquifer.

Arsenic concentrations were found to be declining with tubewell age, although the concentration pattern is uneven and erratic over different time periods (Fig. 6b). Tubewells were first installed in the study area in 1950 and there were only six tubewells prior to the 1971 LiberationWar. The figure increased to 375 by mid 2001. No tubewells were found to be safe that had been installed prior to 1981. The very low negative association ($r = -0.209$) shows a slight decreasing pattern of As concentrations over time. The fitted line with 95 % confidence interval shows a declining trend of As concentrations over time. In addition, the adjusted mean smoothed lowess curve (bandwidth 0.8) shows a declining pattern of relationship between tubewell age and As concentrations. The lowess smoothed line also shows an erratic pattern for recently installed tubewells (Fig. 7b). Tubewell installation year is not a proxy of As concentration dynamics over time since the different concentrations could be location-dependent and not just time-dependent. This can be assessed only within the framework of a monitoring program aimed to assess As concentration in time for given

wells. In this paper As concentration is measured only at one point in time, so it is hard to come to conclusive statements in terms of temporal dynamics.

Discussion

The IK prediction map shows a highly irregular and diverse pattern of As concentration in the study area. A line of evidence shows very high uneven groundwater As concentrations over space.[45–46] About half (46 %) and more than one-fourth (28 %) of the analyzed tubewells measured by atomic fluorescence spectrometry were found to be contaminated with As having the WHO permissible limit and the BDWS respectively and found variation with spatial characteristics having both small-scale variability and large-scale trends.[47] In addition, it was found that almost half of the surveyed 6000 tubewells in Araihazar failed the BDWS for As.[4]

Evidence shows that deep aquifers are almost free from As, but in Bangladesh, only 1 % of tubewells deeper than 200 m have As above 0.05 mg/L and only 5 % exceeded 0.1 mg/L of As.[47] Arsenic is also commonly absent in wells at shallow depths (<5 m), especially in dug wells.[48] In our study, we found that 75 % of the deep tubewells failed to comply WHO standard. At a depth of 12–24 m, themaximumnumber of tubewells was found to be concentrated with As frequently exceeding the BDWS.[4] Harvey et al.[45] showed in their study that dissolved As has a distinct peak at approximately 30 m depth; while Jakariya et al.[13] show a robust association between As concentrations and aquifer depth between 30 and 76 m. A number of studies show typical depth profiles of As concentrations to be "bell-shaped".[4,49–50] Depth trends can be considered within different geologic regions and Yu et al.[3] show a statistically significant trend of decreasing As with depth in different geologic regions of Bangladesh.

Our study shows that As concentration is higher in the fine to medium sediments of the study site. Concurrently, Ahmed et al.[51] and Burges and Ahmed[48] show that As source is preferentially concentrated within fine-grained sediments; while Jakariya et al.[13] argue that a high As concentration is visible at aquifer levelswith the dominance of medium-sized sediments. However, Sharif et al.[8] claim that medium- to coarse-grained aquifer sands are generally less heterogeneous and have a spatially uniform As content. Moreover, Harvey et al.[49] and Swartz et al.[52] note that no chemical characteristics of the solid sediment could be found to explain the high variation of As concentrations with aquifer depth. Spatial variability of groundwater As concentrations is influenced by lithologic heterogeneity, which is controlled by sediment geochemistry, recharge potential, thickness of surface aquitard, local flow dynamics, and the degree of reducing conditions in the aquifer.[8] This incongruity in the relation between grain size and As concentrations might result from the textural properties of the aquifer sands at different depths and aquifer chemistry. It should be noted that a tubewell presently As-free or having an acceptable As concentration cannot be relied upon for long because of the paradoxical nature of concentrations in different space-time dimensions. The literature shows that older wells have a higher probability of high concentrations of As in water. A BGS/DPHE[47] report indicates that those tubewells shallower than 150 m, which contain As at >0.05 mg/L increases consistently with time. A general trend of increasing As concentration with the duration of pumping at individual tubewells was also noticed.[53–55] Nath et al.[56] demonstrate a decreasing trend of As concentration with depth, except for a slight increase observed between 70 and 80 m depth. However, deeper groundwater (>80 m depth) is not entirely free from As. This contradicts the previously reported As distribution scenario in the Bengal Delta Plain.[47,57] These observations imply that As is not released to groundwater uniformly with depth.[58] The

potential for a deep water supply depends on aquifer depth and the lateral extent of a substantial aquitard and its vertical profiles of permeability, coherence, and occurrence of aquitard.

Conclusion

Analysis of the groundwater As concentrations in the study area reveals the significant spatial variability of As. The IK prediction map shows low As concentrations in the north, central and south part of the study area in a scattered manner. The west and northeast of the study area are generally more contaminated, while the southwest part of the study area is contaminated in a highly irregular pattern. The calculated nugget effects, ranges and sills have shown locally erratic variations of As concentrations in the study area. The study has also investigated the deviating relationships between aquifer depths and As concentrations. A low negative association was found for aquifer depths and As concentrations. Deep tubewells were also found to be contaminated, but at a low level of concentration. Associations of As with lithology have also been examined over small regional scales and indicate widespread occurrences of As in the alluvial aquifers that are controlled by larger geological and hydrogeological features.

The spatial pattern of As concentrations in the study area does not show any uniformity corresponding to surface geology. This unevenness in As concentration is due to the aquifer characteristics and surface geology. The study shows thatGanges-deltaic flood plain deposits contain very high and erratic concentrations of As. The sharp microlevel variation of As concentrations in the same aquifer raises a number of issues. Is geological variability the main cause of the differences? The variation ofAs concentrations over time also raises the issue of the mechanism of As in Bangladesh groundwater.

What are the reasons for the variation of As concentration with tubewell age: heavy withdrawal of groundwater for irrigation and domestic uses, or according to geological origin and lithology? Therefore, more research is needed on spatio-temporal analysis, more specifically depth-specific distribution in different geological settings to investigate the nature of As mobilization. Moreover, there is a need to investigate the role of organic matter in the mechanism of As release in groundwater.

Acknowledgments

The senior author would like to express his sincere thanks to Professor Dipankar Chakraborti of the SOES, Jadavpur University, Kolkata, India for his cooperation in the laboratory analysis of the water samples. In addition, we are grateful to three anonymous referees for their constructive remarks.

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