

ARSENIC-SAFE AQUIFERS IN COASTAL BANGLADESH: AN INVESTIGATION WITH ORDINARY KRIGING ESTIMATION

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ABSTRACT:

Spatial point pattern is one of the most suitable methods for analysing groundwater arsenic concentrations. Groundwater arsenic poisoning in Bangladesh has been one of the biggest environmental health disasters in recent times. About 85 million people are exposed to arsenic more than 50µg/L in drinking water. The paper seeks to identify the existing suitable aquifers for arsenic-safe drinking water along with “spatial arsenic discontinuity” using GIS-based spatial geostatistical analysis in a small study site (12.69 km²) in the coastal belt of southwest Bangladesh (Dhopakhali union of Bagerhat district). The relevant spatial data were collected with Geographical Positioning Systems (GPS), arsenic data with field testing kits, tubewell attributes with observation and questionnaire survey. Geostatistics with kriging methods can design water quality monitoring in different aquifers with hydrochemical evaluation by spatial mapping. The paper presents the interpolation of the regional estimates of arsenic data for spatial discontinuity mapping with Ordinary Kriging (OK) method that overcomes the areal bias problem for administrative boundary. This paper also demonstrates the suitability of isopleth maps that is easier to read than choropleth maps. The OK method investigated that around 80 percent of the study site are contaminated following the Bangladesh Drinking Water Standards (BDWS) of 50µg/L. The study identified a very few scattered “pockets” of arsenic-safe zone at the shallow aquifer.

1. INTRODUCTION

Water resources are a prerequisite for human development and progress. Groundwater is purportedly the main source of untreated pathogen-free safe drinking water in more than one-third (2.4 billion) of the total population on the globe (WHO, 2015). But Bangladesh has many water-related problems from public health to social science perspectives. It is ironic that so many tubewells installed to provide pathogen-free drinking water are found to be contaminated with toxic levels of arsenic that threaten the health of millions of people in Bangladesh (Hassan and Atkins, 2011). The impact of arsenic poisoning on human health in Bangladesh has been alleged to be the “worst mass poisoning in human history” (Smith *et al.*, 2000).

As a ubiquitous toxicant and carcinogenic element, groundwater arsenic is associated with a wide range of adverse human health effects (Clewell *et al.*, 2016; Kippler *et al.*, 2016; Lin *et al.*, 2013). Chronic exposure to elevated levels of arsenic is associated with substantial increased risk for a wide array of diseases including skin manifestations (Sarma, 2016); cancers of the lung (Sherwood and Lantz, 2016), bladder (Medeiros and Gandolfi, 2016), liver (Lin *et al.*, 2013), skin (Fraser, 2012), and kidney (Hsu *et al.*, 2013); neurological (Fee, 2016); diabetes (Kuo *et al.*, 2015); and cardiovascular (Barchowsky and States, 2016) diseases. The IARC (International Agency for Research on Cancer) classifies inorganic arsenic as a group-1 human carcinogen and associations have been found with lung, bladder, skin, kidney, liver, and prostate cancer (IARC, 2012).

There is a complex pattern of spatial discontinuity of arsenic concentrations in groundwater with differences between neighbouring wells at different scales and changes with aquifer depth (Hassan and Atkins, 2011; Peters and Burkert, 2008). Spatial discontinuity of arsenic concentration has been reported in Bangladesh (Radloff *et al.*, 2017), West Bengal in India (Biswas *et al.*, 2014), China (Cai *et al.*, 2015; Ma *et al.*, 2016), Chianan Plain of Taiwan (Sengupta *et al.*, 2014), Mekong Delta of Vietnam (Wilbers *et al.*, 2014), the southern Pampa of Argentina (Díaz *et al.*, 2016), the Duero River Basin of Spain (Pardo-Igúzquiza *et al.*, 2015), Nova Scotia in Canada (Dummer *et al.*, 2015), Wisconsin in the USA (Luczaj *et al.*, 2015), the Águeda watershed area in Portuguese district of Guarda and the Spanish provinces of Salamanca and Caceres (Antunes *et al.*, 2014), and so on.

Is it safe to drink tubewell water? Which tubewell water is safe from arsenic poisoning? Which aquifer contains arsenic-safe water and where is it? In answering these questions, it requires an investigation for groundwater management and monitoring. The spatial pattern of arsenic discontinuity with GIS-based kriging estimation can be effective in this connection. Geostatistics and GIS (Geographical Information Systems) technologies have been used as a management and decision tool in the spatial discontinuities of groundwater quality as well as groundwater arsenic concentration (Antunes *et al.*, 2014; Delbari *et al.*, 2016; Flanagan *et al.*, 2016). Geostatistics relies on both statistical and mathematical methods to create surfaces for groundwater arsenic concentrations (Liu *et al.*, 2004). GIS, in the same time, is considered as an automated decision-making system with mapping capabilities for the

geographically referenced information (Achour *et al.*, 2005; Berke, 2004; Burrough and McDonnell, 1998) in preparing spatial mapping for investigating the historical and currently existing arsenic situations in groundwater.

In view of increasing concerns to groundwater arsenic poisoning, this paper focuses on the spatial methodological issues to identify the suitable aquifers for arsenic-safe water management along with spatial arsenic concentrations using geostatistics. Geostatistics with kriging methods can design water quality monitoring in different aquifers with hydrochemical evaluation by spatial mapping.

2. DATA AND METHODS

2.1 Spatial data

GIS is an important methodological issue for spatial mapping to investigate the historical and existing situation of arsenic concentrations in the study site. Points, lines and polygon information were collected through extensive field visits with GPS (Model: Garmin GPSMAP 62STC), small-scale map data, and satellite imageries. This GPS has high-sensitivity receiver with the facilities of preloaded base map with topographic features (Hassan, 2015). Apart from geographical location identification, this device has the facilities for automatic routing with electronic compass and barometric altimeter. The relevant information (i.e. land base and facility base information) were then plotted on GIS environment (ArcGIS). The relevant hard-copy map data for mouza (the lowest level administrative unit in Bangladesh with Jurisdiction List number) sheets with the map scale of RF 1:3960 were arranged for the base map. In addition, the position of each tubewell was plotted on the mouza sheets to check the accuracy of the GPS positional data and vice-versa.

2.2 Arsenic and attribute data

Tubewell screening is important priority work for arsenic data collection. Arsenic is toxic and it is a known documented carcinogen. Therefore, an ethical question was raised: which tubewell would be screened and how many? This was a sensitive issue in the context of present arsenic situation in Bangladesh. Arsenic information from all the 1082 tubewell water samples located in Dhopakali *union* in Bagerhat district in south-west coastal Bangladesh were collected and tested with the HACH field-testing kits in 2014. It is noted that we used to collect tubewell water samples and we took a couple of weeks to collect our water samples from all the tubewells and tested them directly from the field. Moreover, tubewell locations with GPS technology were collected, tubewell depth, installation year, users etc. were collected with observation and face-to-face questionnaire surveys. Dhopakali is a disaster-prone area with a population density of 1052/km² (area: 12.69km²). Use of pond and river water for cooking purposes is a common practice and the region is often considered as the diarrhoea-prone area of the country.

2.3 GIS approach

GIS as a comprehensive set of spatial analytical tool used in analysing arsenic concentration since of its mathematical and programming facilities. Spatial analytical capabilities of GIS were used to identify a spatial pattern of arsenic concentrations. The “iso-arseno” value lines were developed to identify the arsenic concentrations which were predicted through

geostatistical approach. In addition, GIS overlay capabilities allow different map data to be combined in determining “suitable sites” for different arsenic-safe water tables. Reclassification allows the transformation of attribute information; it represents the “recoloring” (Aronoff, 1989) of features in the map. Thus, a map of spatial arsenic concentrations may be classified into categories such as “safe zones”, “contaminated zones” or “severely contaminated zones” without reference to any other information.

2.4 Geostatistics and spatial interpolation

A geostatistical approach relies on both statistical and mathematical methods to create surfaces and to assess the uncertainty of predictions for regionalized variables (Bastante *et al.*, 2008; Ghosh and Parial, 2014; Uyan, 2016) and to assess the uncertainty of predictions. Geostatistics represents one of the most powerful procedures for producing contour maps for regionalised variables (Beliaeff & Cochard, 1995; Xu *et al.*, 2005) and, thereby, indicates an appropriate method of prediction. Geostatistical results, using kriging techniques, are efficient when data for variables are distributed normally (Wu *et al.*, 2014, Uyan *et al.*, 2015). Interpolation is the process of estimating the value of parameters at unsampled points from a surrounding set of measurements (Burrough & McDonnell, 1998). When the local variance of sample values is controlled by the relative spatial distribution of these samples, geostatistics can be used for spatial interpolation and point interpolation is significant in GIS operation (Cinnirella *et al.*, 2005) (Figure 1).

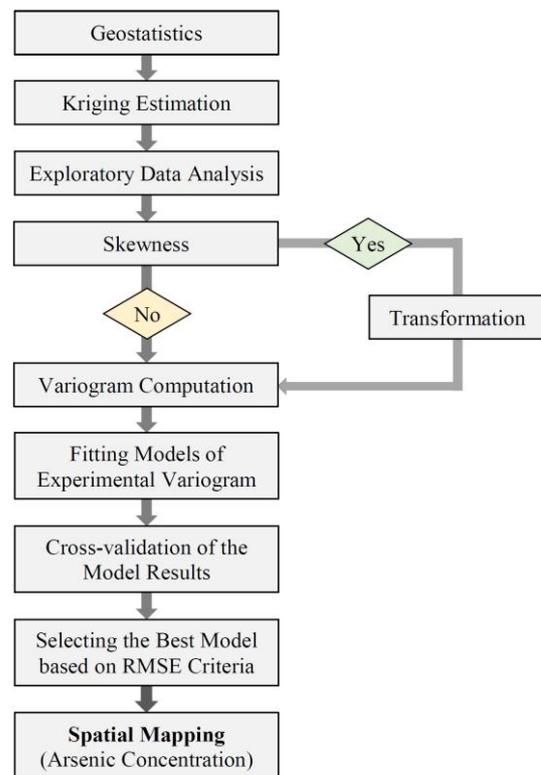


Figure 1. Flow diagram for geostatistical analysis for spatial arsenic concentrations

2.5 Ordinary kriging

Ordinary Kriging (OK) is a geostatistical approach for estimation and linear interpolator that estimates a value at a point of a region for which the variogram is known, without

prior knowledge about the mean of the distribution (Choudhury, 2015; Dokou *et al.*, 2015). In OK, a random function model is used, in which the bias and error variance can both be calculated and then weights are chosen for the nearby samples such that they ensure that the average error for the model is zero and the modelled variance is minimized.

The error variance in OK is based on the configuration of the data and on the variogram, hence is homoscedastic (Yamamoto, 2005). It is not dependent on the data used to make the estimate. Yamamoto (2005) has also shown that the ordinary interpolation variance is a better measure of accuracy of the kriging estimate. OK does not depend on the values of the samples, which means that the same spatial configuration always reproduces the same estimation variance in any part of the area. In order to the estimator to be unbiased in OK, the sum of these weights needs to equal one (Isaaks and Srivastava, 1989). The estimation equation is a linear weighted combination of the form (Journal and Huijbregts, 1978):

$$\hat{Z}(S_0) = \sum_{i=1}^n \lambda_i Z(S_i) \text{ with } \sum_{i=1}^n \lambda_i = 1 \quad \dots \dots \dots (1)$$

OK weights λ_i are allocated to the known values in such that they sum to unity (unbiaseness constraint) and they minimize the kriging estimation variance (Delbari *et al.*, 2016). The weights are determined by solving the following system of equations (Isaaks and Srivastava, 1989):

$$\begin{cases} \sum_{j=1}^n \lambda_j \gamma(x_i, x_j) + \mu = \gamma(x_i, x_0), & i=1, \dots, n \\ \sum_{j=1}^n \lambda_j = 1 \end{cases} \quad \dots \dots \dots (2)$$

Where, $\gamma(x_i, x_j)$ is the average semivariance between pairs of data locations; μ is the Lagrange parameter for the minimization of kriging variance; and $\gamma(x_i, x_0)$ is the average semivariance between the location to be estimated (x_0) and the i th sample point.

OK can be used for spatial pattern of groundwater arsenic concentrations since of its high uneven distribution. OK is used to estimate values when data point values vary or fluctuate around a constant mean value (Serón *et al.*, 2001). It is applied for an unbiased estimate of spatial variation of a component. The estimation variance of OK is used to generate a confidence interval for the corresponding estimate assuming a normal distribution of errors (Goovaerts *et al.*, 2005). The unknown local mean is filtered from the linear estimator by making the sum of kriging weights to one. OK also provides a measure of uncertainty attached to each estimated value through calculating the OK variance (Delbari *et al.*, 2016):

$$\sigma^2(x_0) = \sum_{i=1}^n \lambda_i \gamma(x_i, x_0) + \mu \quad \dots \dots \dots (3)$$

In producing the prediction maps for spatial arsenic concentrations by the OK method, it was specified the semivariogram, search neighbourhood, and nugget model in the interpolation. By using the spherical semivariogram having the nugget value of 7584.601 and the partial sill of 12882.41, with

200 input neighbours (smoothed neighbours, smoothing factor of 0.2) in a neighbourhood shape with anisotropy factor of 1.1692 having 67.67578° axis angle from a test location (X: 89.8602 and Y: 22.7037), the OK prediction map was produced (Figure 2). It is noted that it was also considered the techniques for the validity of the identified arsenic-safe aquifers in the study site.

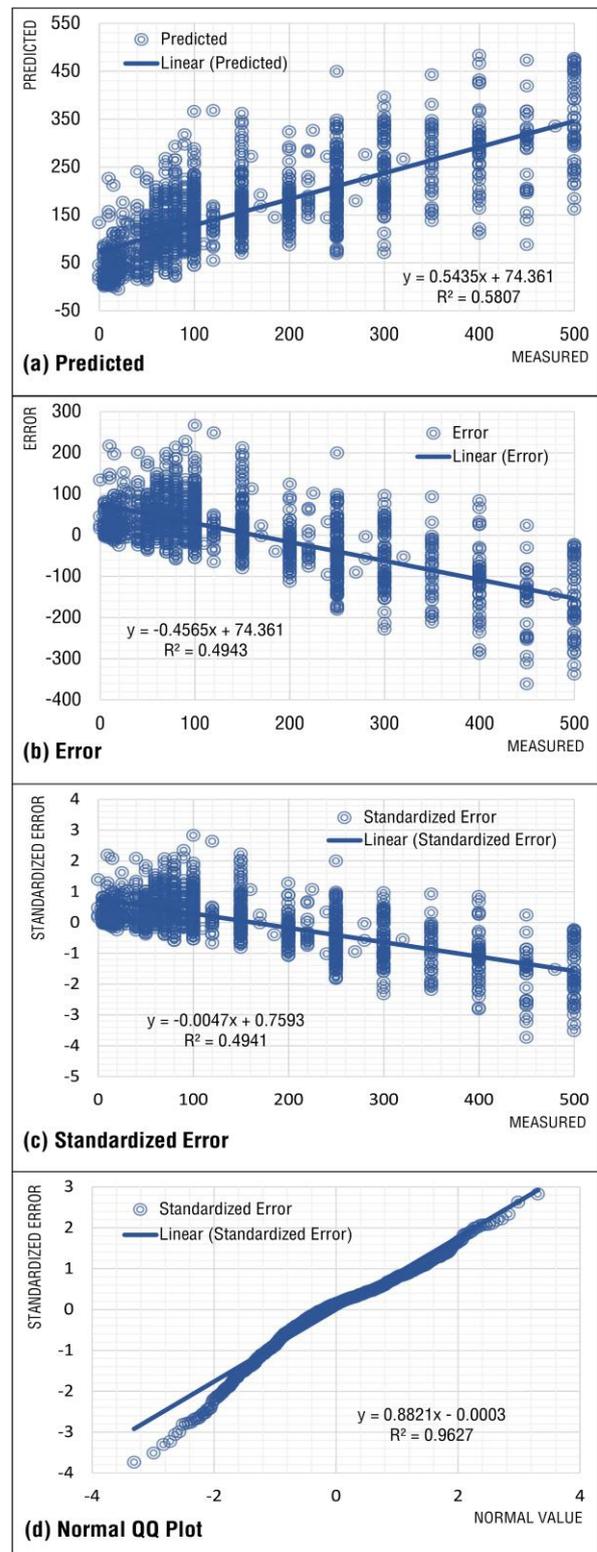


Figure 2. Cross validation with normal trend for the OK prediction map

2.6 Generalized linear models (GLM)

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 (Jin *et al.*, 2005; McCullagh and Nelder, 1989). They are based on an assumed relationship (link function) between the mean of the response variable and the linear combination of the explanatory variables. Since arsenic data are not distributed normally, the GLM was used for this paper. The Newton-Raphson (maximum likelihood) optimization technique is used in this study to estimate the GLM.

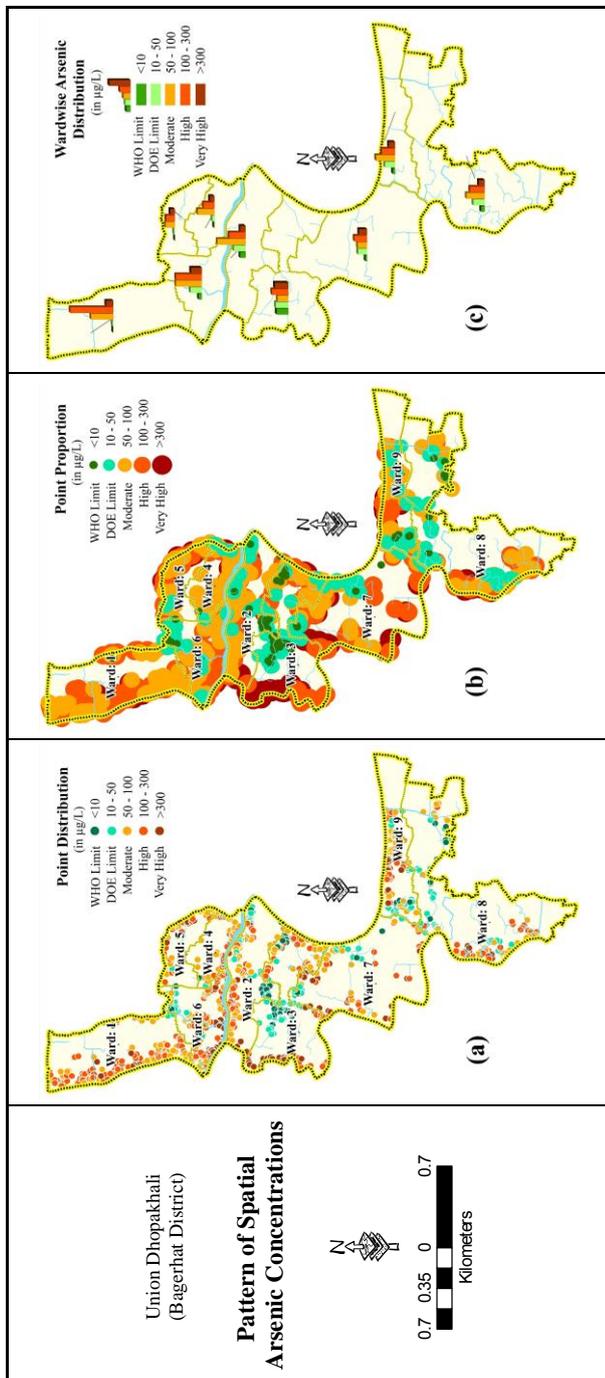


Figure 3. Spatial pattern of arsenic concentrations with different aspects: (a) arsenic in different tubewells; (b) proportion of arsenic concentrations in each tubewell; and (c) pattern of arsenic concentrations at ward level

3. RESULTS AND DISCUSSION

3.1 Arsenic concentration

Arsenic concentrations in groundwater can be classified into different categories based on magnitudes, different permissible limits, and statistical procedures. The term “contamination” in this paper refers to the elevated levels of arsenic concentrations above the Bangladesh Drinking Water Standards (BDWS set by Department of Environment). On the other hand, the “safe level” can be categorized into two different classes: one for WHO permissible limit ($10\mu\text{g/L}$); and another for the BDWS ($50\mu\text{g/L}$). Therefore, arsenic concentrations can be classified into a number of classes (Table 1 and Figure 3): (a) WHO permissible level ($\leq 10\mu\text{g/L}$); (b) BDWS ($10.1-50\mu\text{g/L}$); (c) moderate contamination level ($50.1-100\mu\text{g/L}$); (d) high contamination level ($100.1-300\mu\text{g/L}$); and (e) severe contamination level ($>300\mu\text{g/L}$).

Table 1. Descriptive statistics of arsenic concentrations in Dhopakhalhi Union, Kachua, Bagerhat

Wards	Concentration Level ($\mu\text{g/L}$)					Total Tubewell (Frequency)	Mean ($\mu\text{g/L}$)	Standard Deviation ($\mu\text{g/L}$)
	WHO Limit ($\leq 10\mu\text{g/L}$)	BDWS Limit ($10.1-50\mu\text{g/L}$)	Moderate ($50.1-100\mu\text{g/L}$)	High ($100.1-300\mu\text{g/L}$)	Severe ($>300\mu\text{g/L}$)			
Ward: 1	1	2	49	95	16	163 (15.06%)	209.76	103.04
Ward: 2	12	26	62	36	12	148 (13.68%)	123.99	117.30
Ward: 3	25	28	24	36	30	144 (13.31%)	178.65	178.71
Ward: 4	1	5	37	24	10	77 (7.12%)	168.83	133.74
Ward: 5	-	2	20	19	10	51 (4.71%)	197.84	135.96
Ward: 6	7	25	57	57	24	170 (15.71%)	165.68	128.25
Ward: 7	4	20	24	32	19	99 (9.15%)	181.82	144.48
Ward: 8	8	25	40	32	13	118 (10.91%)	138.43	123.80
Ward: 9	5	22	42	29	14	112 (10.35%)	141.56	127.88
Total:	64 (5.92%)	155 (14.33%)	355 (32.81%)	360 (33.27%)	148 (13.58%)	1082 (100%)	163.01	135.17

Arsenic concentrations are inconsistent with spatial dimension and the pattern of concentrations range $0-500\mu\text{g/L}$, with the mean concentration of $163.008\pm 135.165\mu\text{g/L}$. It was calculated a slight more than one-fifth of the total functional tubewell (20.25%) were found to be safe following the BDWS limit; while almost four-fifth (79.75%) of the tubewell were analyzed with arsenic contamination from moderate to severe levels, while only 5.92% of the total tubewell were analyzed for arsenic-safe following the WHO permissible limit of $10\mu\text{g/L}$.

Groundwater arsenic concentrations were found to be different in each administrative ward in Dhopakali. Elevated levels of arsenic were found in all the administrative wards, but highest mean concentrations were found in Ward 1 (209.76µg/L) followed by Ward 5 (197.84µg/L), Ward 7 (181.82µg/L), and Ward 3 (178.65µg/L) (Table 1). On the contrary, the lowest mean concentration was recorded in Ward 2 (123.99µg/L) followed by Ward 8 (138.43µg/L), Ward 9 (141.56µg/L), Ward 6 (165.68µg/L) and son on. It is noted that mean arsenic concentrations in all the administrative wards are much higher than that of BDWS limit (Table 1 and Figure 3).

There is no deep tubewell (DTW) in Dhopakali and all the functional tubewells are within the shallow aquifer with 10-70m depth. About two-third of the analysed tubewell (704, 65.065%) were installed within 20m depth and they were analyzed with high arsenic contamination, with mean concentration of $152.978 \pm 130.128 \mu\text{g/L}$. Moreover, some 378 tubewell were installed in depths more than 20 meters and mean arsenic concentration were analyzed with $180.648 \pm 141.956 \mu\text{g/L}$.

3.2 Spatial arsenic discontinuity

Which areas are safe and which areas are contaminated? The answer of this question can be analysed with spatial GIS analytical capabilities. The OK prediction method shows the interpolation maps of estimated arsenic concentrations in Dhopakali (Figure 4). A point-in-polygon operation through OK method was performed to analyse spatial arsenic concentrations. In producing the prediction maps, it was specified the power function and search neighbourhood in the interpolation.

Almost one-fifth of the study site are found to be contaminated with elevated levels of arsenic and they are concentrated all over the area except in some parts of the southern and middle of the area. The higher magnitudes are recognizable in the northwest to southwest parts of the study area (Figure 4a). The safe areas identified in the OK estimation are especially in Wards 2 and 3 and the total safe zones cover about 4.17% (53 hectares) of the total study area (Figure 4a). A slight more than one-fifth (20.24%) of the tubewell (219 out of 1082) conform to this safe level.

High and severe contamination zones cover about 51.48% (653 hectares) of the study area; while moderate contamination zones cover about 44.35% (563 hectares). It is noteworthy that the mean arsenic concentration in Dhopakali is more than three times higher (163.01µg/L) than the BDWS (50µg/L) and more than 16 times higher than the WHO permissible limit (10µg/L). Moreover, arsenic concentrations were found to be high erratic with aquifer depth (Figure 4bc).

The pattern of arsenic concentration varies considerably and unpredictably over distances of a few meters. In the study area, about 71% of tubewell are located within 43 meters of each other, but within this distance there are remarkable variations. The overall pattern of arsenic concentrations in groundwater within the settlement area in Dhopakali shows a moderate contamination running along the banks of the Taleshwar River to the central part of the area. Safe zones are mainly concentrated in the central and south-eastern part of the study area in a scattered manner (Figure 4); while the contaminated zones are concentrated into the west and north-western parts of the study area. The contaminated zones are found everywhere in the study area but with a decrease in the degree of

contamination from west to east. In addition, areas close to river bank are generally more contaminated; while the south-eastern parts of the study area are contaminated in a highly irregular pattern (Figure 4).

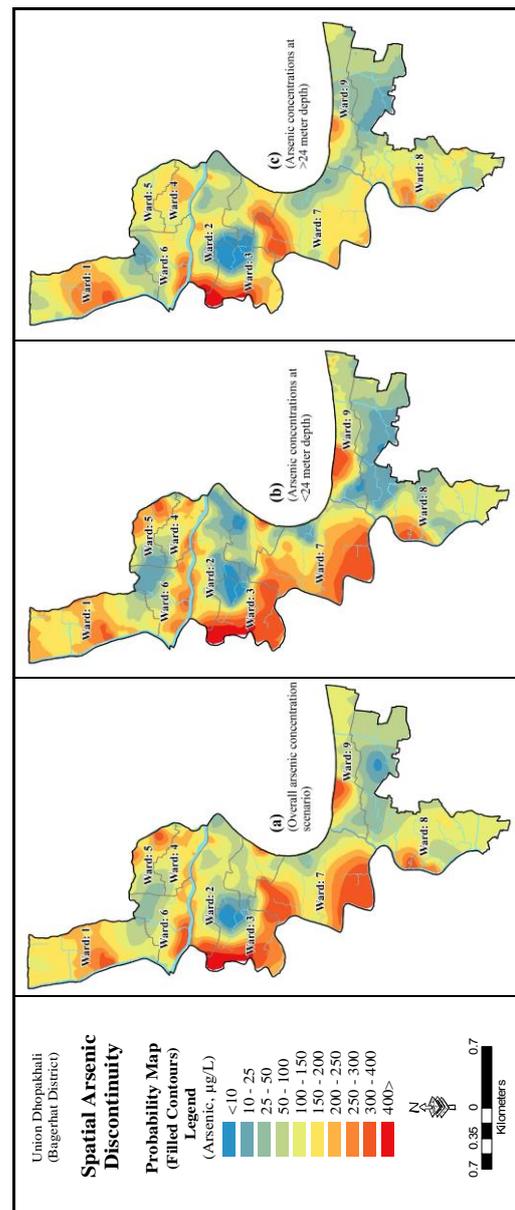


Figure 4. Spatial arsenic discontinuity: (a) Overall scenario in the study site; (b) at the depth less than 24 meters; and (c) at the depth more than 24 meters

3.3 Safe water demand areas

Which areas will get priority in getting access to safe drinking water? The answer of this question can detect the safe-water “command areas” and safe-water “demand areas”. Assurance of drinking-water safety is a foundation for prevention and control of waterborne diseases. We have already identified safe and contaminated areas following the concentration levels of toxic inorganic arsenic with OK approach. A very small number of areas were identified as safe zones. The safe water command areas were identified in Wards 2 and 3 in Dhopakali and the total safe zones cover about 12.53% (49 hectares) of acreage within the settlement area in Dhopakali.

The safe water command areas are located in some parts of the south-eastern and the middle of Dhopakali, but very few safe tubewells are located in irregular pattern in other administrative Wards (Figure 4). A slightly more than one-fifth (20.24%) of the tubewells (219 out of 1082) have been identified within the arsenic-safe zones. It is estimated that people who are living within the high and severe contamination zones are needed for safe-water options and the tubewell technology is not suitable in the contaminated areas. About 87.47% (342 hectares) of the settlement area are within the unsafe zones. There is no DTW in Dhopakali and shallow tubewell (STW) are not suitable in the identified demand areas. It is noted that installation of more STW is not required as urgent for safe-water command areas.

3.4 Suitable area for safe tubewell installation

Identification of suitable arsenic-safe aquifer is an important objective for this study. Suitability analysis is a process of systematically identifying or rating potential locations with respect to a particular use. The OK approach has identified the spatial determination for suitable areas for tubewell installation with aquifer depths and water-tables (Figure 5).

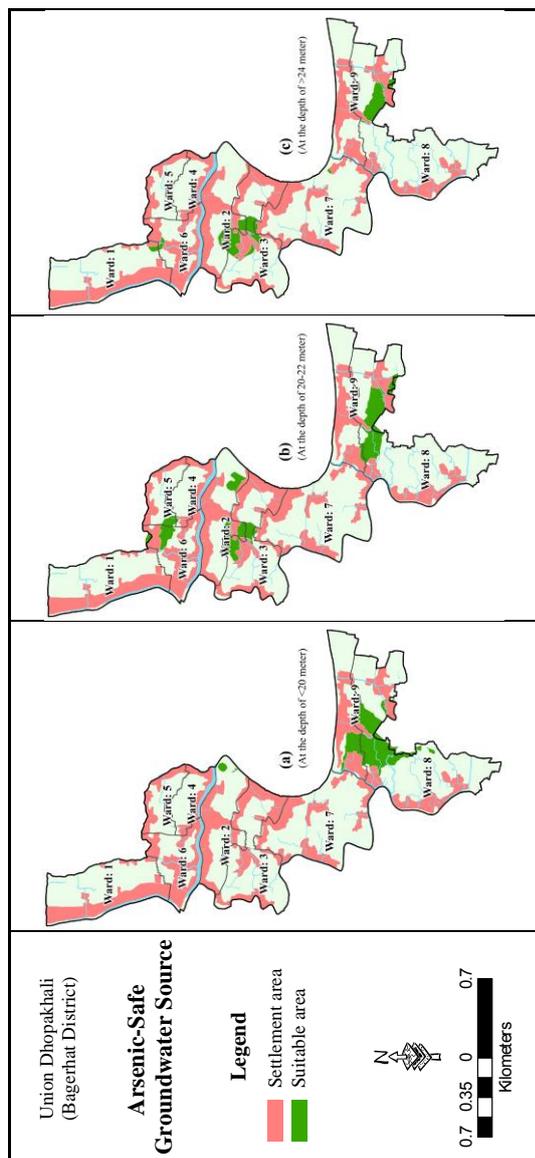


Figure 5. Suitable arsenic-safe aquifer in Dhopakali

It is noted that only the technological option for both the STW and DTW was considered for this Spatial Decision-Support System (SDSS). In considering the safe-water “command areas” and safe-water “demand areas”, we tried to identify the suitable areas for installing tubewell in different aquifer depths. Accordingly, we identified places suitable for installing tubewell for arsenic-safe water (Figure 5).

Existing arsenic-safe “command areas” in Dhopakali has been identified following the safe concentration levels of arsenic in STW. Apart from STW, people are habituated untreated pond water for their drinking and cooking purposes in Dhopakali. We didn’t consider this water source for safe-water “command areas” - we have considered only shallow aquifer for suitable area identification for safe-water through tubewell option. Figure 5 shows the suitable areas for arsenic-safe water at shallow aquifer. We have classified the aquifer based on water table and they were categorized as: (a) lower than 20m depth; (b) 20-22m depth; and (c) more than 22m depth (Table 3). At the depth of <20 meters, there are suitable areas for arsenic-safe STW option for installation more precisely and a number of settlement clusters in Wards 2, 3, 7, 8, and 9 with sporadic distribution pattern (Figure 5). At the depth of 22-22 meter, there is an arsenic-safe water table and it is distributed in all the administrative Wards except Ward 5 (Figure 5).

It is noted that the identified STW suitable areas are mainly located in the north-eastern, central, and south-eastern parts of the study area (Figure 5). Moreover, at the depth of >22 meter, arsenic-safe water can be tapping mainly from Ward 3, but there are very small areas for arsenic-safe water and they are in Wards 1, 2, and 6 (Figure 5 and Table 3). We have identified from our fieldworks that the sub-surface geology in Dhopakali is not suitable for installing DTW. Moreover, the deep aquifer is heavily concentrated with sodium chloride.

Table 3 shows the arsenic-safe suitable aquifer in the study area. The areas have been identified at the micro level. People can easily locate which sites are best fitted for getting arsenic-safe water at which depth. Moreover, this planning can be helpful for future strategic plan to provide alternative technological option in providing safe drinking water.

Depth (meter)	Wards	Village	Settlement Cluster (Para)	
<20	Ward: 2	Masokhali	Purbo Para	
	Ward: 3	Barodaria	Purbo Para	
	Ward: 7	Vasa	Uttar Para, Haldar Para	
	Ward: 8	Shanpukuria	Paschim Para, Dakshin Para	
		Sherokhali	Dakshin Para	
	Ward: 9	Vasa	Purbo Para	
		Boga	Purbo Para	
	20-22	Ward: 1	Kamarganti	Purbo Para
		Ward: 2	Masokhali	Dakshin Para
Ward: 3		Barodaria	Purbo Para	
		Boyarsinga	Purbo Para	
Ward: 4		Madhovkathi	Hawladar Bari	
Ward: 6		Jhalodanga	Moddho Para, Hazi Bari	
		Sitabari	Uttar Para	
Ward: 7		Vasa	Uttar Para	
		Sherokhali	Dakshin Para	
Ward: 8	Shanpukuria	Paschim Para		
	Vasa	Maddho Para, Purbo Para		
Ward: 9	Boga	Paschim Para		

>22	Ward: 1	Kamarganti	Purbo Para
	Ward: 2	Masokhali	Dakshin Para, East Para
	Ward: 3	Barodaria	Purbo Para
	Ward: 6	Jhalodanga	Hazi Bari

Table 3. Suitable area for arsenic-safe tubewell installation in the study site

4. CONCLUSIONS

The study has mainly attempt to investigate the spatial pattern of groundwater arsenic concentrations and to identify suitable areas for installing STW, the most acceptable water technology. Mapping the proximity area of arsenic and spatial GIS overlay capabilities allow different map data to be combined in determining suitable sites for different arsenic-safe water tables. Based on the existing arsenic information and characteristics of water tables, it was demarcated the right areas for arsenic-safe water at different aquifer depths in the study area. Considering the situation of groundwater, it can be taken a decision that further installation of DTW would not be significant for safe drinking water.

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