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STATISTICAL EVALUATION OF HIGHLY ARSENIC CONTAMINATED GROUNDWATER IN SOUTH-WESTERN BANGLADESH

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Abstract: *High Arsenic (As) in natural groundwater in most of the shallow sandy aquifers of the South-Western part of Bangladesh has recently been focused as a serious environmental concern. This paper is aiming to illustrate the statistical evaluation of the Arsenic polluted groundwater to identify the correlation of that As with other participating groundwater parameters so that the As contamination level can easily be predicted by analyzing only those parameters. Multivariate data analysis done with the collected groundwaters from the 67 tube-wells of the contaminated aquifer suggests that As may have substantial positive correlations with Fe, Mn, Al, DOC, HCO_3 and PO_4 whereas noticeable negative relationships have also been observed with SO_4, Cl and NO_3-N. Based on these relationships, a multiple linear regression model has been developed that incorporates seven most influential groundwater parameters as the independent predictor variables to estimate the As contamination level in the polluted groundwater. This model could also be a suggestive tool while designing the As removal scheme for the affected groundwater.*

Key words: *groundwater; arsenic; multiple regression; principal component; prediction*

1. Introduction

Arsenic a toxic, trace element is ubiquitous in nature [1][5]. It can easily be transported from the sediment to the surrounding pore-water. In the subsurface environment, As (arsenic) has generally been occurring in naturally driven sediments. Partitioning of As in the sediment-water interface has been controlled by a variety of geochemical factors [3,5]. When this As gets exposed in the groundwater in a higher concentrations, it then can pose a threat to the public health. Presence of As in excessive amount may also cause several skin diseases that may lead to cancer eventually [1].

Due to the easier accessibility coupled with having pathogen free nature, groundwater (GW) extraction promoted by the UNICEF had been thought as the proven resources for providing drinking water to the Bangladeshi villagers, who had shifted from the surface water sources to get rid of the water borne diseases over the past 30 years [5]. However, recent discoveries of high Arsenic contamination in GW put about 35 million people forward to a severe health risk [1,2,3,4,5,6,7]. Such type of high As concentration in the natural groundwater, has also been recognized as the serious public health problems in the other parts of the Southern Asia such as West Bengal (India), Vietnam, Cambodia, Taiwan, China and Inner Mongolia over the years [1]. In Bangladesh, since early 1990s, As pollution observed in the upper shallow groundwater was mostly reported in the Holocene deltaic and alluvial sedimentary aquifers [3,4,5,6,8]. The national survey conducted by the BGS and DPHE in 2001, stated that around 27% of the total 10 million shallow tube-wells have exceeded the safe drinking water limit of the WHO (10 μg/l) and the Bangladesh guide line value (50 μg/l) [3,7,8].

This ground water As distribution pattern appears to follow the regional geology [10]. Since the Pleistocene and Holocene period, sediments carried by the Ganges-Brahmaputra-Meghna river system had been consolidating to construct the deltaic aquifer near the coastal region [8, 9]. Silt, clay and fine to medium sand have frequently been observed in this Holocene sediment. Three principal types of aquifers have also been documented in these Holocene sediments depending on the depth, age and water availability. The ground water originated from the unconsolidated fine to finer sediments of the first aquifer (10-100m) has noticeably been contaminated with As [5]. The next aquifer which comprises mostly of fine to medium sand (110-170m) is the second aquifer and the deeper part composing of medium to coarse sand is the third aquifer [12]. This second and third aquifers have generally been reported to have lower As concentrations. Presence of impervious clay layers is the dominating factor that may act as the separator between two adjacent aquifers. This also restrict vertical As mobilization from upper to the underneath aquifers.

South-Western part of Bangladesh predominantly, is the worst hit region where almost 70% of the existing shallow tube-wells exceed the drinking water standard (50μg/l). Samples of groundwater were collected from one of the Arsenic hot spots located at a village named as Alaipur (22^0 51' 30.4" N, 89^0 03 ' 41.2 "E, around 300 km from capital city Dhaka) under Kolaroa upazilla of Satkhira district of South-Western Bangladesh. The topographic surface of that area is very flat and the land elevation with respect to the mean sea level does not exceed even 5 m [5]. The reason behind the selection of this affected area for this study is that several people of this area have already been identified to be badly infected with Arsenicosis diseases. Moreover, as that community is far from the capital city, that area has still been remaining unexplored.

According to the borehole investigations of the sub-surface geology, brown clay layers in the upper shallow part predominately were observed to have most of the detectable As. Just below this layer fine grey reduced sandy layers containing significant amount of As were also found up to a depth of 40 m where most of the shallow hand tube wells have been screened.

Slow aquifer flushing rate coupled with the relatively longer residence time have triggered the Arsenic problem in the alluvial aquifers [3]. Arsenic that may be leached from the upper source layer, mobilizes towards the surrounding groundwater and gets mixed after rearranging the equilibrium of the groundwater system.

Over the years, several researchers have been attempting to explore the As problems in various aspects. However, few of them have worked to present the correlation model of As with other groundwater ions. The purpose of this paper is to present the statistically correlated groundwater parameters that may have either positive or adverse influences on As mobilization and also to show the As concentration predictive model developed with these influential parameters by using multiple regression technique. This model will apparently help to efficiently carry out the As removal design from the contaminated groundwaters.

Multivariate technique in treating environmental data has long been practiced in assessing spatial or temporal groundwater contamination caused either by natural or anthropogenic factors. Cluster analysis (CA) and principal component analysis (PCA) are the two important tools of the multivariate techniques that are applied to determine the dominant interrelationships among the variables to understand the processes that may responsible in controlling the groundwater chemistry [7,13]. Multiple linear regression [14] model has also been applied in evaluating the level of an emerging groundwater contaminant that may have correlationship with other chemical elements [2]. Multiple linear regression (MLR) technique can further help to predict the level of a promising respondent pollutant by showing the relative influences of the other employed predictor pollutants.

2. Materials and Methods

Fifteen monitoring wells were installed by following local drilling practice such as percussion method. This method includes a systematic and synchronized action of continuous raising and lowering of a steel pipe attached with a lever and having a diamond cutter at its mouth. Apart from this, 52 other surrounding tubewells were selected for sampling of Arsenic contaminated groundwaters. Groundwater samples (GW) from these tubewells were collected using peristaltic pumps that were maintained a nearly constant flow rate of 200 ml/min along with an attached multi probe flow channel accommodating pH, DO (dissolved oxygen), ORP (Eh, redox potential) and EC (electrical conductivity) meters' electrodes in the flow line. Water samples from each well were collected in three different ways for serving different purposes such as filtered (0.45 μm filter) for examining anions, unfiltered for dissolved organic carbon (DOC) analysis and filtered and then acidified samples (ultra pure HNO_3) for analyzing major cations. Groundwater samples collected (GW) from the selected locations and depths of the targeted aquifer were subjected to carry out the aqueous phase analysis [11]. ICP-MS (Inductively Coupled Plasma Mass Spectrometry) and IC (Ion chromatography) were employed to do the aqueous analysis for determining major cations such as As (arsenic), Fe (iron), Mn (manganese), Al (aluminum) and anions such as PO_4 (phosphate), SO_4 (sulfate), Cl (chloride), NO_3-N (nitrate nitrogen), DOC (dissolved organic carbon) and HCO_3 (bicarbonate). Multivariate Data analysis (MVDA) technique was applied to find out the relationship among the participating ions.

The raw data were treated first with z-scale transformation to make the data standardized. Multivariate data analysis (MVDA) was utilized to identify the correlationship among the measured groundwater parameters. Principal component analysis was done to reduce the number of input variables. Spearman's correlation matrix was performed to illustrate the correlation coefficients among the variables. Finally, multiple linear regression (MLR) technique was employed to develop a model that can be applied to realize the level of As contamination by predicting the groundwater As from the other measured parameters. Chemometrics software and SAS were explored to perform the multivariate data analysis and also to develop multiple linear regression model.

3. Results and Discussions

3.1. Groundwater Data

The basic statistics of the geochemical data for the analyzed groundwater samples are presented in the Table A-1 (Appendix A). The maximum As concentration was found markedly in the upper shallow aquifer as 0.180 mg/L whereas minimum of that was reported in the comparatively deeper layer as only 0.010 mg/L. Groundwater having high As concentration was also reported to contain the high concentrations of Fe (12.3 mg/L), Al (0.48 mg/L), Mn (0.067 mg/L), DOC (6.87 mg/L), PO_4 (0.13 mg/L) and HCO_3 (508 mg/L). However, low concentrations of Cl (59 mg/L), SO_4 (0.89 mg/L) and NO_3-N (1.23 mg/L) were observed to be associated with the groundwater carrying elevated As. Moreover, this groundwater was noticed to have very low Eh (redox potential, 5 mV) which may indicate the reductive nature of the aquifer geo-environment where the As has always been mobilizing after being continuously leached from the sediment matrix. The strong correlations reportedly found among the parameters such as Fe, Al, Mn and As in the contaminated groundwater further suggest that these elements may have been releasing from the common source minerals in the sediments [5]. High Ca (calcium) concentration in As affected groundwater was also noticed remarkably in most of the samples.

DOC and HCO_3 that usually evolve with the potential microbial activities occurred in the younger sediments may also have positive influences on As, since As is supposed to be released concurrently with the Fe-oxide dissolution. PO_4 on the other hand, may appear in the aquifer groundwater either from the dissolution of the phosphate minerals or from the leaching of the used chemical fertilizers. The prevailing competition for searching of adsorption sites that is presumed to be occurred among the participating ions including As, PO_4 and HCO_3 may also enhance As mobilization. In addition, low sulphate content in high As affected groundwater reflects the likely occurrence of the sulphate reduction that might have a considerable contribution in mobilizing As in the aquifer. However, Cl has been noticed to have adverse influence on As. It has overwhelmingly been observed that due to the presence of elevated Cl concentrations, As content in the groundwater has significantly been found lower. Such type of finding has also been reported for the groundwater identified with high Na (Sodium) and EC (electrical conductivity) content. Low nitrate content was also another observation of the high As groundwater. The dominant pH was in the near neutral range. Na, Mg, K are the parameters that usually reflect either the evidences of the likely intrusion of the sea water into the aquifers or the presence of saline water trapped in the aquifer.

3.2. Loading Plot of Principal Component Analysis

Two principal components P1 and P2 explain notably the variances of the nature and influences of the selected variables. The loading plot of the principal component analysis portrays very well the correlations of the groundwater parameters in the Fig.1.

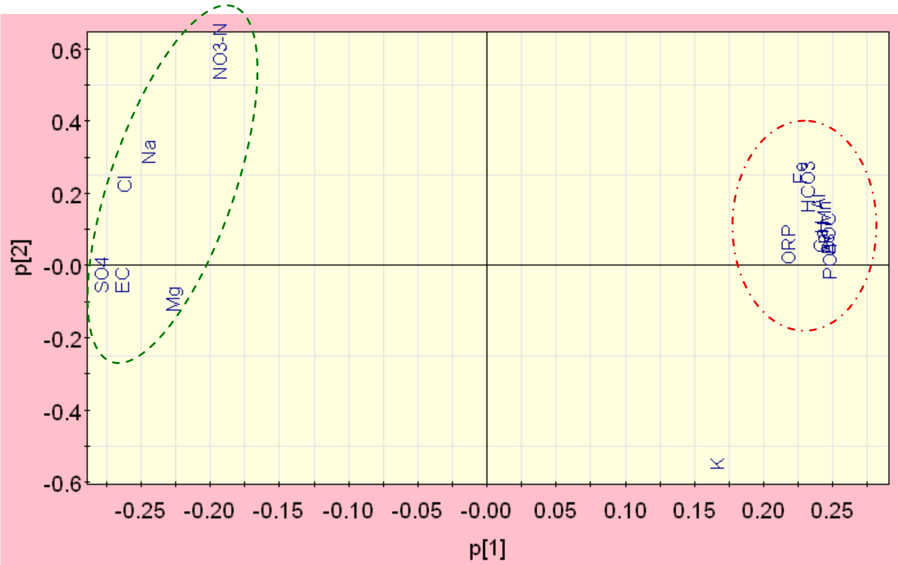

Figure1. Loading plot of the principal component analysis of the studied groundwater.

Parameters having positive influences on As have noticeably been found to make clusters among themselves. This strong relationship of As with other parameters such as Fe, Mn, Al, PO_4, HCO_3, and DOC can explicitly be visualized in the right circle of the loading plot (Fig. 1). Adversely influencing parameters particularly SO_4, Cl and NO_3, on the other hand, can also be recognized in the left circle of the loading plot (Fig. 1). It seems that K may have insignificant relationship with As, since this is placed dispersedly in the diagram. These findings are very much consistent with the bivariate analysis that can be found elsewhere [3,5].

3.3. Non parametric Correlationship of the GW Parameters

The non parametric Spearman correlation analysis was accomplished to identify the plausible statistical relationships that may exist among the observed As concentrations and other groundwater variables: Fe, Al, Mn, DOC, HCO_3, PO_4, SO_4, NO_3-N and Cl. The Spearman correlation coefficient usually presents the strength of the relationship that may exist between any two considered parameters by indicating either positive or negative magnitude.

The Spearman correlation matrix illustrated in Table A-2 (Appendix-A), shows the high correlation coefficients for As with Fe, Mn, Al, PO_4, HCO_3 and DOC. However, lower coefficients for Cl, NO_3-N and SO_4 with As are also listed in that Table. This coefficient matrix strongly supports the observations that were found in the loading plot analysis.

3.4. Hierarchical Cluster Analysis

Plot of hierarchical cluster analysis done with Ward's mode incorporating Euclidean distance, for the groundwater parameters is portrayed in Fig. 2. Showing the As, Mn and Al in the same cluster, this plot again reflect that the stronger correlations may exist among these parameters. Positions of SO_4 and NO_3-N in the same cluster may also reflect their very much distinguishable characteristics in compare to the other parameters. Fe here is seen to be very closely linked with DOC and this may also indicate the possible role of organic material in accomplishing the reductive dissolution of iron minerals which may again be considered to control the release of As by initiating the co-dissolution of the attached As compound [3,8].

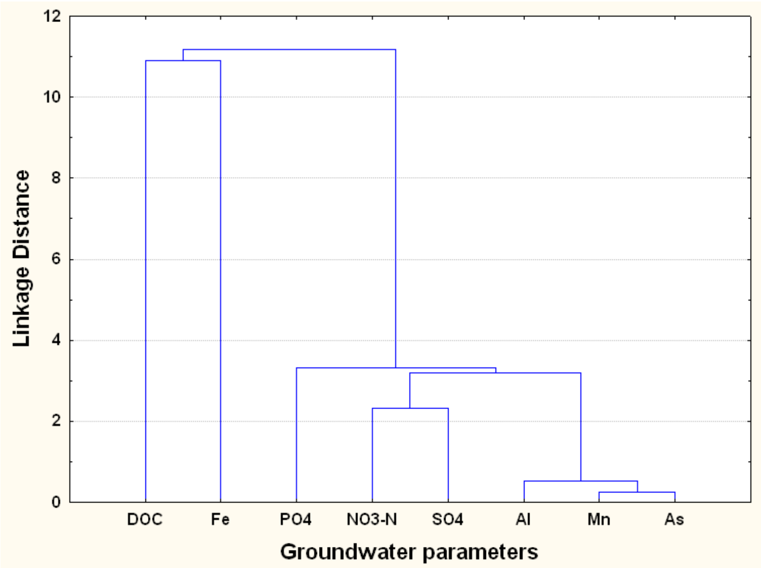

Figure 2. Hierarchical cluster analysis plot of the groundwater parameters

3.5 Multiple Linear Regression Model

Generally a multiple regression analysis attempts to fit the independent variables for predicting a single dependent variable. The general form of a model developed with multiple regression looks like [14]:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \ldots\ldots\ldots + \varepsilon \quad (1)$$

Where,
X_1, X_2, X_3 denote the independent variables,
Y stands for the dependent variable,
β_0, β_1, β_2, β_3 represent the correlation coefficients,
and ε designates the error term.

The seven selected independent variables such as Fe, Mn, Al, PO_4, DOC, Cl and NO_3-N and one independent variable As were used as the input data for fitting the multiple regression model as shown in the equation no. 1. SAS was employed to develop the model. The results of the model are presented in Table 1 and Table 2.

Table 1. Analysis of variance

Source	Degree of Freedom	Sum of Squares	Mean Square	F value	p-value
Model	7	0.05661626	0.006291	10.404	<0.0001
Error	60	0.00003674	6.123E-6		
Corrected Total	67	0.05665300			

Table 2. Parameter estimation

Parameters	Estimate	Std Error	t-value	p-value
Intercept	-0.602392	0.355907	1.69	0.1415
Al	0.052438	0.056234	0.93	0.00011
Fe	0.0023208	0.00146	1.59	0.0029
Mn	0.1370394	0.565053	0.24	0.00015
DOC	0.0257569	0.006016	4.28	0.00032
PO_4	0.0030028	0.032017	0.09	0.00042
Cl	-0.000019	1.114E-5	1.71	0.00013
NO_3-N	-0.01882	0.006178	3.05	0.0026

Based on the analyzed parameters listed in the Table 2, the final model can be formulated as:

$$As = 0.052\ Al + 0.0023\ Fe + 0.137\ Mn + 0.025\ DOC + 0.003\ PO_4 - 0.000019\ Cl - 0.019\ NO_3\text{-}N$$

This model may suggest the prediction of the As contamination by measuring the seven predictor parameters of the groundwater variables in any contaminated aquifer. This model may also be a suggestive tool in predicting As contamination level while designing the As removal activities by the Environmental scientists.

4. Conclusion

Various statistical analysis techniques have successfully been applied in this study to evaluate the geochemical phenomena of the highly Arsenic affected groundwater in the shallow sandy aquifers of the South-Western Bangladesh. It has markedly been observed here that the As in that aquifer sediment has gradually been releasing with the great influence of the naturally driven geochemical factors. HCO_3 and DOC have been recognized as the two most responsible geochemical parameters that may have controlled As releasing mechanism. These two parameters may also have influenced the Fe, Mn and Al source minerals to initiate their dissolution reactions under the reductive geo-environment.

Loading plot coupled with the hierarchical plot has also presented either the possible relationships that may exist among the groundwater parameters. Positive influences of Fe, Mn, Al, PO_4, DOC and HCO_3 and adverse influences of Cl and NO_3-N on groundwater As that have been observed in the multivariate analysis as well as in the Spearman rank table, are also consistent with the Arsenic releasing as well as the mobilizing phenomena.

A predictor model, developed with seven dominant independent parameters of groundwater to perceive an estimation of the Arsenic contamination level, has been briefly portrayed in this study. This model may further guide the Environmental Scientists to design an efficient Arsenic removal plan while treating the groundwater that may have contaminated badly with that.

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Appendix A

Table A-1. Basic statistics of the GW parameters (mg/L)

Parameter	Mean	Minimum	Maximum	Std. Dev.
As	0.088	0.002	0.205	0.062
Al	0.193	0.010	0.520	0.146
Ca	57.700	12.300	114.200	33.204
Fe	5.603	1.010	16.200	4.184
K	2.621	1.030	4.500	1.024
Mg	25.626	14.700	36.230	6.560
Mn	0.039	0.010	0.081	0.022
Na	114.636	12.700	513.000	158.084
pH	6.881	6.590	7.290	0.191
EC	1609.125	886.000	2510.000	598.917
ORP	4.250	-7.000	45.000	23.621
DOC	4.354	1.670	7.650	1.862
PO_4	1.906	1.023	2.670	0.474
SO_4	0.855	0.310	1.420	0.320
As	0.088	0.002	0.205	0.066
Cl	452.750	31.000	1501.000	560.917
HCO_3	288.875	187.000	492.000	84.883
NO_3-N	1.386	1.070	1.900	0.254

Table A-2. Spearman correlation matrix for the groundwater parameters

	As	Fe	Al	HCO_3	Cl	DOC	Mn	PO_4	SO_4	NO_3-N
As	1.000									
Fe	0.610	1.000								
Al	0.524	0.517	1.000							
HCO_3	0.697	0.409	0.570	1.000						
Cl	0.072	0.053	-0.020	0.037	1.000					
DOC	0.468	0.559	0.291	0.172	0.125	1.000				
Mn	0.418	0.506	0.503	0.418	-0.038	0.367	1.000			
PO_4	0.394	0.244	0.415	0.609	-0.102	0.155	0464	1.000		
SO_4	-0.096	-0.132	-0.198	-0.351	-0.483	-0.026	-0.309	-0.187	1.000	
NO_3-N	-0.541	-0.293	-0.398	-0.411	-0.024	-0.414	-0.377	-0.225	0.034	1.000

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