

Factor and Cluster Analysis of Water Quality Data of the Groundwater Wells of Kushtia, Bangladesh: Implication for Arsenic Enrichment and Mobilization

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Abstract: The study area is located in the southwestern part of Bangladesh. Twenty-six groundwater samples were collected from both shallow and deep tube wells ranging in depth from 20 to 60 m. Multivariate statistical analyses including factor analysis, cluster analysis and multidimensional scaling were applied to the hydrogeochemical data. The results show that a few factors adequately represent the traits that define water chemistry. The first factor of Fe and HCO_3 is strongly influenced by bacterial Fe (III) reduction which would raise both Fe and HCO_3 concentrations in water. Na, Cl, Ca, Mg and PO_4 are grouped under the second factor representing the salinity sources of waters. The third factor, represented by As, Mn, SO_4 and K is related to As mobilization processes. Cluster analysis has been applied for the interpretation of the groundwater quality data. Initially Piper methods have been employed to obtain a first idea on the water types in the study area. Hierarchical cluster analysis was carried out for further classification of water types in the study area. Twelve components, namely, pH, Fe, Mn, As, Ca, Mg, Na, K, HCO_3 , Cl, SO_4 and NO_3 have been used for this purpose. With hierarchical clustering analysis the water samples have been classified into 3 clusters. They are very high, high and moderately As-enriched groundwater as well as groundwater with elevated SO_4 .

Keywords: Piper, Hierarchical Cluster analysis, Groundwater, Arsenic, Bangladesh.

INTRODUCTION

Groundwater As-enrichment in Bangladesh is considered to be one of the worst environmental disasters in the world (Ahmed et al., 2004). Tens of millions of Bengali and Bangladeshi people are at health risk from exposure to As-enriched groundwater (BGS and DPHE, 2001). Arsenic is derived from the weathering of Himalayan rocks in tectonically active areas. Released As is strongly adsorbed by stream sediments and oxides under aerobic conditions when it is transported by surface water. Sediments with sorbed As are then deposited with organic matter in alluvial settings. Subsequently, Fe(III)-reducing bacteria present in alluvial sediments cause the release of sorbed As to groundwater under moderately reducing conditions (Saunders et al. 2005; Polizzotto et al. 2008). The two most plausible mechanisms for the release of As from sediments to groundwater are oxidation of sulfides (Chatterjee et al. 1995) and reduction of Fe- and Mn-oxyhydroxides (Bhattacharya et al. 1997, 2004; Nickson et al. 1998). The former hypothesis suggests that As-rich Fe sulfides may be dissolved by oxidation, driven in part by increased

groundwater pumping. The latter hypothesis implies the reduction of As-rich Fe-oxyhydroxides, caused by increased O_2 levels possibly associated with human activities, or buried peat and organic deposits (McArthur et al. 2001). Hydrochemists have applied factor analysis to interpret observed relationships among variables, to yield statistical relationships that provide insight into the underlying structure of the variables, to assess controls on chemical composition of groundwater and to evaluate the spatial distribution of the studied components (Matalas and Reier, 1967). Statistical analyses have indicated that As, Fe, Mn, Ca and PO_4 are strongly correlated in the Holocene alluvial sediments in the Manikganj district of Bangladesh (Shamsudduha et al., 2008). Liu et al. (2003) adopted factor analysis to identify the factors that govern the chemistry of groundwater in the southern Choushui river alluvial fan, and demonstrated that the hydrogeochemical process of As-enrichment is the main characteristic of this groundwater. Wang et al. (2007) applied factor analysis to identify the factors that govern the chemistry of groundwater in two groundwater catchments in Taiwan. Groundwater is an

economic resource, apart from drinking purposes, it is used in agriculture, industries and municipalities, and water obtained from underground sources should be chemically pure and reasonably free from microorganisms. According to a UNICEF study, access to safe drinking water in Bangladesh has declined by 17% in the last three years due to arsenic contamination. An understanding of both the quality and quantity of groundwater is needed when evaluating groundwater resources. Chemical analysis of major cations and anions in groundwater is helpful in determining its suitability for agricultural, industrial and domestic purposes (Karim et al. 2002). The quality of water when in its physical, chemical, and biological characteristics deteriorates and becomes harmful to man, aquatic life or to a balanced functioning of the aquatic ecosystem, then it is said to be polluted or contaminated. The study area is of great concern and is located in the southwestern part of Bangladesh. It lies between $23^{\circ}21'30''N$ to $24^{\circ}8'30''N$ latitude and $88^{\circ}42'30''E$ to $89^{\circ}21'30''E$ longitude (Fig.1). The total area of the Khustia Municipality is approximately 120 sq. km. It is bounded in the north by Natore and Pabna district, in the east by Rajbari District, in the south by Meherpur, Chuadanga and Jhenaidah, and in the west by India. The area is well connected with Dhaka and Rajshahi and its adjoining areas both by road and rail. There are a number of roads in the study area. The study area falls under subtropical monsoon climate with three main seasons in a year viz. winter, pre-monsoon and monsoon. Winter, which is quite pleasant, prevails from November to February when the temperature ranges from $9^{\circ}C$ to $15^{\circ}C$. This season is cool, dry, and almost rainless. Temperature starts rising from February and reaches a maximum of $38.5^{\circ}C$ in the month of April and May. The mean monthly relative humidity varies from 60 to 88%. Maximum and minimum humidity is observed in the month of July and March respectively. The average monthly maximum and minimum evaporation 110 mm and 40 mm is observed in the months of April and December respectively. The soils of the study area are developed over unconsolidated alluvial sediments. The area may be divided into two agricultural types viz. floodplain soil and soil of Marshes and Beels. Common floodplain feature is the

Beel. Several Beels and marshes are present in the study area and most of them dry up during summer. The elevation of the area ranges from 8 to 14 m above mean sea level (MSL).

The important rivers in the area are mainly Padma, Gorai, Kumar and Kaliganga. Gorai, Kumar and Kaliganga Rivers are not permanent. The drainage pattern is dendritic, typical of all older Pleistocene terraces in Bangladesh (Reiman, 1993). During rainy season these rivers act as excellent drainage channels, draining off a large volume of water and have a considerable current. Most of these rivers are narrow and flow along well-defined channels. Quaternary sediments of the study area can be subdivided into two broad geologic groups, such as (a) Pleistocene older alluvium and (b) Holocene sediments. The surface water system of the study area is associated with the river Padma and its flood plain streams, beels, canals, ponds and rainwater, which is temporarily retained on the land surface until its secondary movements as overland runoff or infiltration towards the sub-surface. Both rivers and beels provide a major source of surface water. The irrigation system of Khustia Municipal area depends both on rainwater and on groundwater. The aim of this study is to determine the As mobilization process and the factors responsible for the elevated As level in the alluvial aquifers in the southwestern part of Bangladesh.

MATERIALS AND METHODS

Twenty-six groundwater samples were collected from

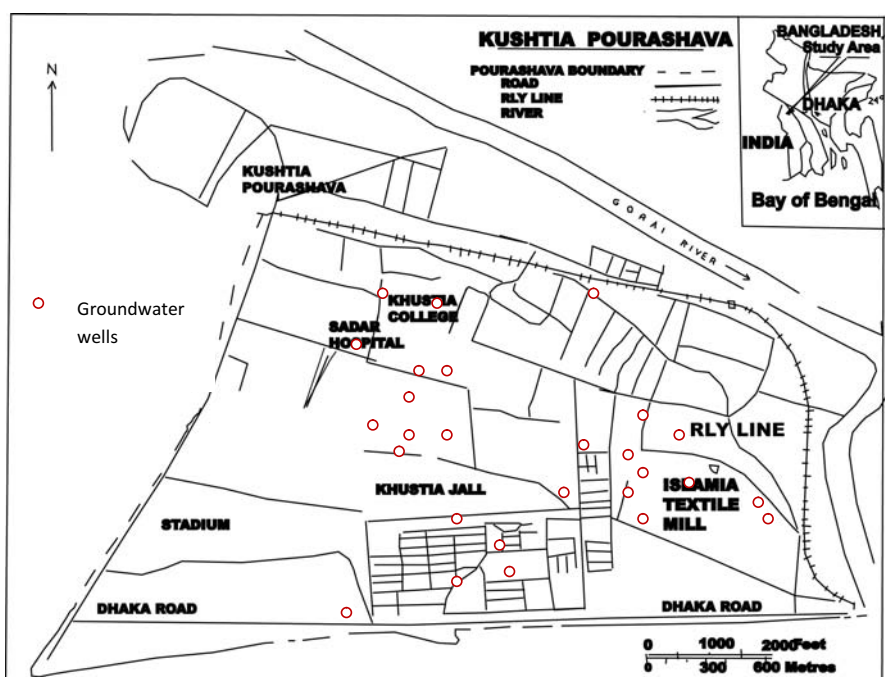


Fig.1. Location map of the study area.

shallow and deep tube wells ranging in depth from 20 to 60 m, for major ion and trace element analyses. At least three well-volumes of groundwater were pumped before sampling. Groundwater samples for trace element and major cation analyses were filtered through 0.45 µm mesh size acetate–cellulose membrane filters and acidulated with HNO₃. Samples were delivered to the laboratory within 24 h. Anions such as NO₃⁻, SO₄²⁻ and Cl⁻ (Table 1) were determined by ion chromatography (Dionex, CA, USA). The errors of ion chromatography for anion analyses are <1%. Cations and trace elements were measured using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent CE 7500, Japan). The errors of ICP-MS for all trace element analyses are <1%. Groundwater samples including blank, spike, duplicate, and check samples (standard solutions from Merck) were measured sequentially. Piper diagrams developed by the Waterloo groundwater software were used for the classification of water types. The software package SPSS version 15 (SPSS Inc., 1998) was utilized for statistical analyses. The techniques used include factor analysis, hierarchical cluster analysis and multidimensional scaling methods. Cluster analysis is a method for placing objects into more or less homogeneous groups so that the relation between the groups is revealed. The general procedure for hierarchical clustering is described in detail in Fig. 2. Hierarchical clustering joins similar observations and then successively connects the next similar observations to these. First the matrix of similarities between all pairs of observations is calculated. Those pairs having the highest similarities are then merged, and the matrix is recomputed. This is done by averaging similarities that the combined observations have with other observations. The progression of levels of similarity at which observation merge is displayed as dendrogram.

RESULTS AND DISCUSSIONS

The major ion composition of groundwater samples from Kushtia district is presented in Table 1. The major cations

Table 1. Summary of the range and average of chemical composition of groundwater samples (n = 26) from Kushtia of northwestern Bangladesh

Parameters	Maximum	Minimum	Average	Standard deviation
As (mg/L)	1.08	0.01	0.37	0.43
Fe (mg/L)	3.00	0.03	0.97	0.63
Mn (mg/L)	1.52	0.02	0.53	0.33
Na (mg/L)	38.50	11.60	26.87	6.60
K (mg/L)	8.70	1.50	4.73	1.65
Ca (mg/L)	148.50	55.50	88.81	21.56
Mg (mg/L)	55.25	10.68	33.54	14.90
Cl (mg/L)	86.50	35.00	64.09	13.29
HCO ₃ (mg/L)	540.50	255.40	388.48	68.34
PO ₄ (mg/L)	10.00	2.40	5.03	1.70
SO ₄ (mg/L)	6.00	1.00	3.19	1.52

in the sampled groundwater are Ca (46–14 mg/L) and Na (11–38 mg/L). Bicarbonate (200–540 mg/L) is the major anion in the groundwater of the study area, whereas Cl (35–175 mg/L) is the second dominant anion. Groundwater SO₄ concentrations ranges from 1 to 6 mg/L. Major ion composition plotted on a Piper diagram indicates that the groundwater is mostly Ca–HCO₃ type in the Kushtia district (Fig. 3). Concentrations of dissolved As, Fe, and Mn in groundwater of Kushtia area are given in Table 1. Mean concentration of As in 26 water samples from tube wells in Chapai-Nawabganj is 350 µg/L. The maximum As level is 1080 µg/L, which is almost 108 times higher than the WHO standard.

Iron is another important trace element commonly found in higher concentrations in Bangladesh groundwater. The average Fe concentration in groundwater is 990µg/L (Table 1). The maximum Fe concentration is approximately 3000µg/L and the minimum is 30µg/L. Manganese concentrations range from 20 to 1520 µg/L with a mean value of 500 µg/L. Groundwaters with high As concentrations contain relatively high concentrations of Fe and Mn also in most cases, but are generally low in dissolved SO₄ concentrations. This relationship is consistent with other parts of Bangladesh where, groundwater contains high

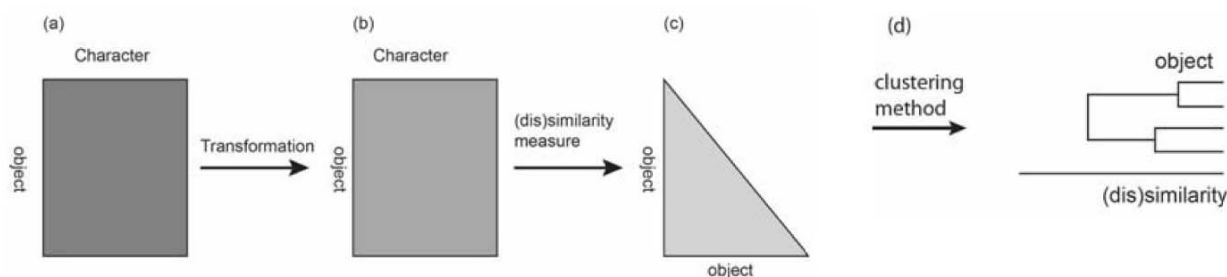


Fig.2.The general procedure for hierarchical clustering.

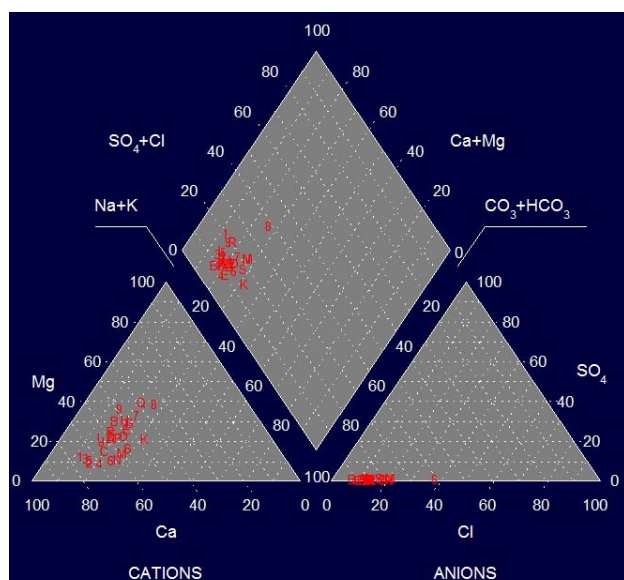


Fig.3. Piper diagram.

concentrations of As (BGS and DPHE, 2001). Several multivariate statistical analyses (Glynn and Plummer, 2005) were undertaken to examine the relationship between As and other groundwater constituents. Pearson's correlation analysis was performed on multivariate components of Kushtia groundwater on their log-transformed values. Eleven variables (Fe, Mn, As, Ca, Mg, Na, K, HCO_3^- , Cl, SO_4^{2-} , and PO_4^{3-}) in groundwater from 26 wells were selected for factor analysis. The major components of the three principal factors were deduced (Table 2). The first factor consists of Fe and HCO_3^- ; the second factor covers Na, Cl, Ca, Mg and PO_4^{3-} and the third factor is composed of As, Mn, SO_4^{2-} , and K. When the factor analysis was completed, an attempt to interpret the principal factors was undertaken.

Factor 1, having the principal components Fe and HCO_3^- , points to the carbonate producing reactions involving the bacterial breakdown of organic matter under reducing conditions. The factor can conveniently be termed as the reduction factor. Factor 1, which has strong positive and

negative loadings on Fe and HCO_3^- , respectively, explains 18.94% of the total variance. The presence of dissolved carbonate may promote the desorption of As and trace metals by competing for sites on mineral surfaces. The free carbonate or carbonate complexes formed may react with reduced Fe and Mn ions to form precipitates such as siderite and rhodochrosite (Villalobos et al. 2001). Harvey et al. (2002) indicated that the dissolved organic C (DOC) and dissolved inorganic C (DIC) in groundwater in Bangladesh were positively correlated with As content, and concluded that the mobility of As is closely related to the recent inflow of C or desorption of As by carbonate or detrital organic C. Factor 1 points to the dissolution of carbonate minerals. The factor can also conveniently be termed as the hardness factor.

Factor 2, which has strong positive loadings on Na, Cl, Ca, Mg and PO_4^{3-} explains 17.75% of the total variance. The strong positive terms are the dominant solutes in seawater. Thus, Factor 2 is called the salinization factor. This factor represents the main dissolved load of groundwaters as a result of the interaction with minerals or trapped saline fluids in aquifers and chemical weathering of catchment rocks.

Factor 3, dominated by As and Mn, could be associated with enrichment of As and can, therefore, be referred to as the As-enrichment factor. Arsenic correlates poorly with Fe, whereas, As is positively correlated with Mn in groundwater. A perfect correlation between As and Mn does not exist because the precipitation of rhodochrosite (Mn carbonate) solids tends to remove some dissolved Mn (but not As) from groundwater. The poor correlation between As and Fe may be caused by the removal of Fe as FeCO_3 solids (Nickson et al. 2000; Lee et al. 2007). It may be concluded that As could be released into groundwater due to reductive dissolution of $\text{MnO}(\text{OH})$ or $\text{FeO}(\text{OH})$ as bacteria oxidizes organic matter to gain energy (Anawar et al., 2003; Ohno et al., 2005; Shamsudduha et al., 2008). The experimental results confirmed that the bacterial dissolution of Fe and Mn appeared to occur with the dissolution of As and that the main mechanism involves the dissimilatory reductions of Fe(III), Mn(IV) and As(V) due to the presence of indigenous bacteria (Lee et al. 2010). The statistical results of this study also agree with the experimental results.

Arsenic can be immobilized via its sorption onto Fe- and Mn-oxides. It has been reported that As seems to be remobilized by the dissolution of these oxides due to microbial reduction of Fe and Mn and that this process appears to be ubiquitous in anoxic environments (Lovley, 1993; Nealson and Saffarini, 1994; Lee et al. 2005). The relationship between As and Mn is regarded as involving the release of As under reducing condition and thus Factor 3 (16.75% of the total variance) can be termed 'the arsenic

Table 2. Rotated Component Matrix

	1	2	3
As	-0.358	0.267	0.617
Fe	0.877	0.035	0.013
Mn	0.149	0.039	0.439
Na	-0.128	0.758	0.082
K	-0.259	0.069	0.753
Ca	0.408	0.549	-0.101
Mg	0.074	0.639	0.116
Cl	-0.398	0.442	0.272
HCO_3^-	0.766	0.099	0.090
PO_4^{3-}	0.328	0.618	-0.536
SO_4^{2-}	0.238	-0.055	0.550

mobilization factor'. Anawar et al. (2002) reported that most of the groundwater samples contain high concentrations of As, dissolved Fe, Mn, NH₄, PO₄ and HCO₃, and low concentrations of SO₄ and NO₃ reflecting the reducing condition of the groundwater. Arsenic is also found to be associated with Fe, Mn, Si, Ba and pH in many other groundwater aquifers in Bangladesh and West Bengal, India (BGS and DPHE, 2001; Dowling et al. 2002; Stüben et al. 2003; Hasan et al. 2007). These observations support that bacterial reduction processes would release aqueous As, Fe, Mn, and other trace metals such as Sr and Ba that have been adsorbed or co-precipitated by oxides or other silicate minerals.

Figure 3 shows the results of plotting the 26 water samples (STW1-STW20 and DTW2-DTW5) on the Piper diagram. The chemical types of groundwater of the study area are distinguished as groups by their position on the Piper diagram. The data is broadly distributed rather than forming distinct clusters. Chemical differences in the distribution of major ions are clearly visible in the Piper diagram. The diagram shows that the distribution of cations in water samples is mainly characterized by Ca ions. The distribution of anions is dominated by HCO₃. The mean concentrations of the analyzed components for each cluster reveals that cluster 1, is highly enriched in As (mean 430 µg/ L) (Table 3), cluster 2 is moderately enriched with As (mean 300 µg/ L) and cluster 3 is highly enriched with As (mean 530 µg/L) with high SO₄ content may be due to anthropogenic influence. Detailed methodologies have been developed for cluster analyses and results have been evaluated. The first step is the selection of the proper data set. The total 26 well samples have been used for the

cluster analysis. The parameters chosen for the clustering were the 12 major components pH, As, Fe, Mn, Ca, Mg, Na, K, Cl, SO₄, HCO₃ and NO₃. The selected data is believed to be most adequate for the cluster analysis. Subsequently, the hierarchical cluster analysis technique has to be selected, the transformation of data should be considered and the dissimilarity matrices computed. The hierarchical cluster analysis has been selected for an adequate partitioning of the water samples into water types or water groups. The dissimilarity matrices of the analysed components of the well samples have been computed using the squared Euclidian distance method. The method is available in the SPSS software package and is found to be well suited to analyse the data of the wells.

Finally, the clustering itself can be performed and the dendrograms have to be prepared and analysed. The clustering can also be carried out with the SPSS code using multi-variate statistics and the resulting dendrograms should be printed on paper. Preparation of the dendrograms on the computer screen would have been an option. With the hierarchical cluster analysis 3 clusters have been distinguished for the well data. The dendrogram of the clusters is shown in Fig. 4. The dendrogram shows the structure of the main clusters of the analysed water samples. Table 3 shows the mean values of chemical constituents for each cluster. Multidimensional scaling (MDS) was used to visualize the dissimilarity matrix and the results of the cluster analysis. For example, Fig. 5 shows the MDS results of three clusters in two dimensional presentations of point plots with the appropriate delineations. The results of the hydrochemical interpretation of the clusters of the water are summarized as follows:

Cluster 1 (CaHCO₃): This is highly mineralised hard water with very high levels of Ca and HCO₃. Arsenic concentration is high in groundwater of the study area. Possibly reductive dissolution of MnO(OH) is the mechanism for release of arsenic from sediment into groundwater. Elevated Ca, Mg and HCO₃ concentrations over Na and Cl is caused by the interaction of groundwater with aquifer sediments rich in carbonate and organic matter (Ahmed et al. 2004). This groundwater generally has low SO₄ content. In the analyses, cluster 1 covered a water type (quite abundant) that was characterized as mainly anaerobic and low in dissolved solids. The water is moderately anaerobic with very high As, Fe and Mn concentrations. Mn concentration is high indicating the reducing environment.

Cluster 2 (CaHCO₃): This water type has high total dissolved solids content, which is mainly due to its high hardness indicated by high values for calcium bicarbonate.

Table 3. Mean values of chemical constituents for each cluster (all units are in mg/L except pH)

Parameter	Cluster 1	Cluster 2	Cluster 3
Wells	STW-5, 6,9,13, 15,10,19,20 DTW-5	STW-16, 17,18, 2,1,7,11,14,12 DTW-7, 6,2,3,4	STW-3, 4,8
pH	7.61	8.10	7.33
Na	30.51	23.84	30.11
K	4.62	4.77	4.90
Ca	109.18	75.43	90.13
Mg	36.33	33.28	26.40
Cl	62.78	61.48	80.17
HCO ₃	391.47	406.47	295.52
SO ₄	3.00	3.07	4.33
NO ₃	1.23	0.99	0.43
As	0.43	0.30	0.53
Mn	1.10	0.98	0.54
Fe	0.48	0.57	0.54
Major Hydrochemistry	CaHCO ₃	CaHCO ₃	CaHCO ₃

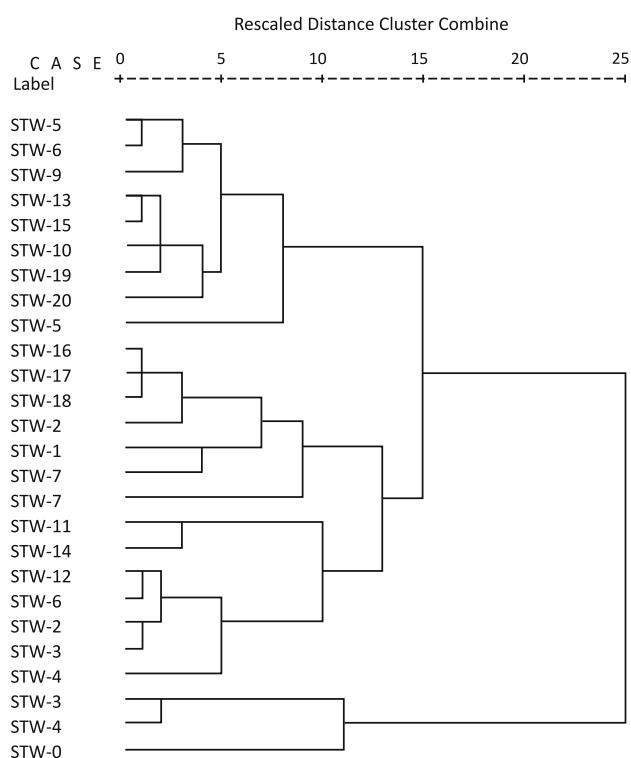


Fig.4. Dendrogram for cluster structure.

The other features of this water are that it is highly anaerobic, indicated by the high iron concentrations. A carbonate buffered groundwater type is suggested by the high CaHCO_3 concentration. Samples from this type are mainly found in carbonate containing sediments. The water types are primarily a result of the dissolution of carbonate minerals, whereby surface water charged with atmospheric and biogenic CO_2 infiltrates into the subsurface. However, this particular limewater type is anaerobic, not influenced by agricultural activities. Most likely, this water is influenced by the infiltration of surface water. The water needs standard treatment and softening treatment due to the high hardness. Iron concentration is high. Arsenic concentration is high in groundwater of the study area. Possibly reductive dissolution

of MnO(OH)/FeO(OH) is the mechanism for release of arsenic from sediment into groundwater.

Cluster 3 (CaHCO_3): This water is mineralized with elevated levels of Ca and HCO_3 levels indicating the lime water type and also distinctive Na, K and Cl levels and anaerobic. The higher Na, K and Cl indicate a certain influence of old marine layers or Holocene marine transgression. The influence of agricultural activities is most likely absent due to low concentration of NO_3 , but has an additional raised chloride concentration. Arsenic concentration is very high in groundwater of the study area. Possibly reductive dissolution of FeO(OH)/MnO(OH) is the mechanism for release of arsenic from sediment into groundwater. The water needs standard treatment and softening because of the high hardness. This water has high levels of total dissolved solids and is mineralized, moderately anaerobic (elevated Fe and Mn) and in balance with respect to carbonate. The higher Na and Cl concentrations indicate that this water may be abstracted from wells that have some influence from marine formations. Sulfate concentrations in the groundwater are relatively high. High SO_4 concentrations may be derived from seawater or caused by pyrite oxidation. The waters can be referred to as a mixed water type (mix of calcite dissolution and influence of old marine layers). The Ca and Mg ions are derived from carbonates (e.g., calcite and dolomite) and high Na and Cl contents suggest a marine source or mixing with seawater. This water can be referred to as a mixed water type (with influence from carbonate dissolution and marine connate water. This is consistent with the report by McArthur et al. (2001).

CONCLUSIONS

Using factor analysis, the chemical constituents were grouped into three main factors on the basis of correlation coefficients and principal components. These are a reduction factor, salinity factor and an As-enrichment factor. Hydrochemical facies analysis showed that most of the water samples are dominated by Ca and HCO_3 ions. The hierarchical clustering analysis indicates that the water from the shallow tube wells can be classified into three clusters based on multivariate statistics. Cluster 3 and 1 water types are highly enriched in As; an alternative water source is thus needed for domestic water supply. Possible solutions are to install tube wells in the deeper Pleistocene aquifers or use clean surface water sources such as reservoirs or rain water. Cluster 2 water types contain comparatively low concentration As but exceeds the Bangladesh standard ($>50 \mu\text{g/L}$), and this type of water is not also suitable for extraction

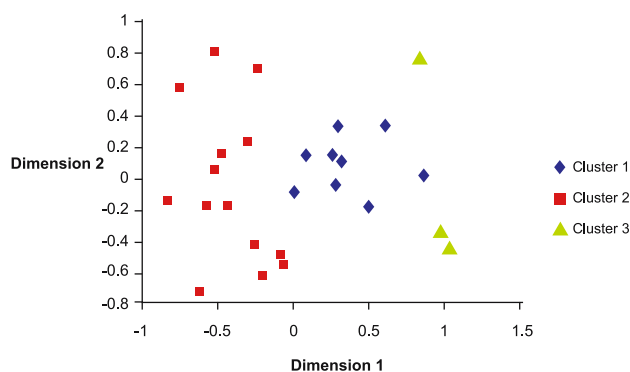


Fig.5. Multidimensional scaling of three clusters.

for domestic uses. Correlation analyses clearly show that As is not correlated with Fe in groundwater but is positively correlated with Mn. These correlations along with results of sequential leaching experiments suggest that reductive dissolution of MnO(OH) and FeO(OH) mediated by

anaerobic bacteria is the primary mechanism for releasing As into the groundwater (Reza et al. 2010). However, the Fe concentrations in groundwater may be modified by precipitation of Fe-mineral phases such as siderite or vivianite under reducing conditions.

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