Water Science

ScienceDirect

Water Science 30 (2016) 19–40

journal homepage: www.elsevier.com/locate/wsj

Characterization of groundwater quality using water evaluation indices, multivariate statistics and geostatistics in central Bangladesh

Md. Bodrud-Doza[a], A.R.M. Towfiqul Islam[b,c], Fahad Ahmed[a], Samiran Das[d], Narottam Saha[e], M. Safiur Rahman[f,g,*]

[a] *Department of Environmental Sciences, Jahangirnagar University, Dhaka 1342, Bangladesh*
[b] *School of Atmospheric Sciences, Nanjing University of Information Science and Technology, Nanjing 210044, China*[1]
[c] *Department of Disaster Management, Begum Rokeya University, Rangpur 5400, Bangladesh*
[d] *School of Hydrometeorology, Nanjing University of Information Science and Technology, China*
[e] *School of Earth Sciences, University of Queensland, Brisbane, Australia*
[f] *Department of Environmental Engineering, Dalhouise University, Halifax, Canada*
[g] *Atmospheric and Environmental Chemistry Laboratory, Chemistry Division, Atomic Energy Center, Dhaka 1000, Bangladesh*[1]

Received 8 August 2015; accepted 10 May 2016
Available online 16 July 2016

Abstract

This study investigates the groundwater quality in the Faridpur district of central Bangladesh based on preselected 60 sample points. Water evaluation indices and a number of statistical approaches such as multivariate statistics and geostatistics are applied to characterize water quality, which is a major factor for controlling the groundwater quality in term of drinking purposes. The study reveal that EC, TDS, Ca^{2+}, total As and Fe values of groundwater samples exceeded Bangladesh and international standards. Ground water quality index (GWQI) exhibited that about 47% of the samples were belonging to good quality water for drinking purposes. The heavy metal pollution index (HPI), degree of contamination (C_d), heavy metal evaluation index (HEI) reveal that most of the samples belong to low level of pollution. However, C_d provide better alternative than other indices. Principle component analysis (PCA) suggests that groundwater quality is mainly related to geogenic (rock–water interaction) and anthropogenic source (agrogenic and domestic sewage) in the study area. Subsequently, the findings of cluster analysis (CA) and correlation matrix (CM) are also consistent with the PCA results. The spatial distributions of groundwater quality parameters are determined by geostatistical modeling. The exponential semivariagram model is validated as the best fitted models for most of the indices values. It is expected that outcomes of the study will provide insights for decision makers taking proper measures for groundwater quality management in central Bangladesh.

Keywords: Geostatistics; Hydrochemistry; Groundwater quality; Water indices; Central Bangladesh

* Corresponding author at: Department of Environmental Engineering, Dalhouise University, Halifax, Canada. Tel.: +1 9024480089.
[1] Present address.
Peer review under responsibility of National Water Research Center.

Production and hosting by Elsevier

http://dx.doi.org/10.1016/j.wsj.2016.05.001

1. Introduction

Sustainable groundwater quality is important for drinking, irrigation and domestic purposes. Groundwater quality has become a major concern in Bangladesh (Shahidullah et al., 2000; Raihan and Alam, 2008; Bahar and Reza, 2010; Rahman et al., 2012a,b; Biswas et al., 2014). The central part of Bangladesh is facing the problem of declining groundwater quality due to several reasons such as shifting of river natural direction, improper management of water body, climatic variability and anthropogenic activities. Arsenic contamination and mobilization into groundwater also affect the water quality in central Bangladesh (BGS and DPHE, 2001).

However, heavy metals contamination is also of great concern on lives owing to their toxicity, persistence and bioaccumulation in central Bangladesh. Continuous monitoring and evaluation of the groundwater quality, thus helps to save lives and environment (McCutcheon et al., 1993; Meharg and Rahman, 2003; Islam et al., 2015). Islam et al. (2015) found that concentrations of Fe and Mn were found higher than other heavy metals in groundwater of Bangladesh. Although, groundwater pollution rate is not so high, groundwater is the only option for good quality water in Bangladesh. Accessibility of drinking water in Bangladesh has increased over the past decade, adverse impact of unsafe drinking water on health continues (WHO, 2004). Foster (1995) investigated that groundwater quality are posing great threat due to intensive use of natural resources and increased human activities (Fig. 1). Evaluation of groundwater quality is a complex process that undertaking numerous variables capable of causing various stresses on general groundwater quality. However, characterization of groundwater quality in central Bangladesh by using integrated appropriate methodologies is not yet to be carried out.

Therefore, this study has been designed to elaborately illustrate an integrated approach that includes drinking water indices, several pollution indices, multivariate statistics and geostatistics modeling to characterize the groundwater quality in the Faridpur district of central Bangladesh. Various researchers have tried to develop a wide range of WQIs for evaluation of groundwater quality; the choice of index depends on the groundwater input parameters and the desired results (Vasanthavigar et al., 2010; Singh et al., 2013; Tiwari et al., 2014; Shahid et al., 2014). For instant, water quality index (WQI) is an effective technique for assessing drinking water quality suitability in any area and to communicate the information on overall water quality. Heavy metal pollution index (HPI), heavy metal evaluation index (HEI) and degree of contamination (C_d) are used to evaluate the hazardous metal pollution in drinking water purposes (Prasad and Bose, 2001; Edet and Offiong, 2002). However, the WQI values have limitation, which cannot provide evidence of the pollution sources. The WQI values, thus have to be used together with heavy metal pollution index (HPI), heavy

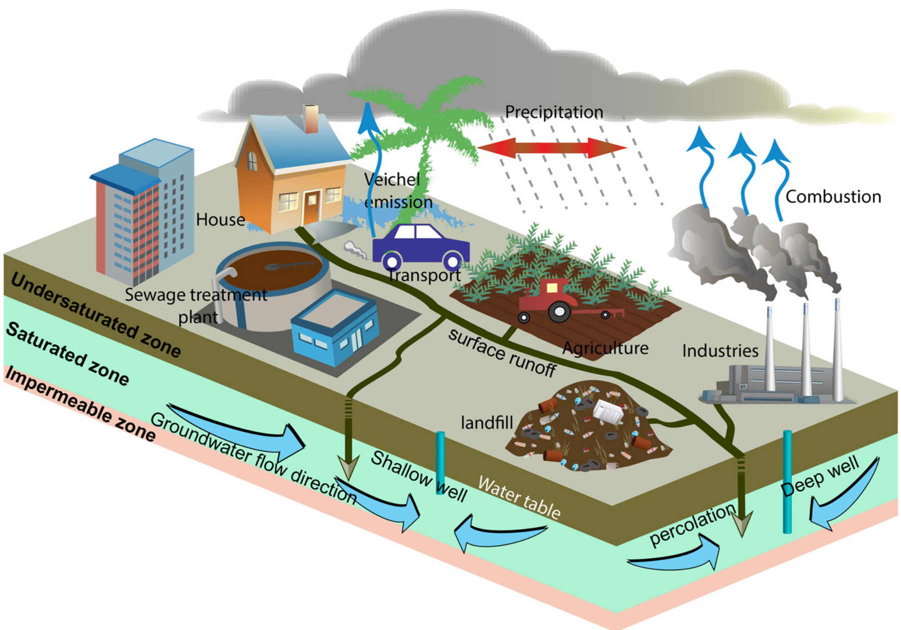

Fig. 1. Groundwater contamination depending on natural and anthropogenic activities.

metal evaluation index (HEI) and degree of contamination (C_d) for better assessment of groundwater pollution levels. So, water quality indices are useful guideline for environmental manager, decision makers, water planner of a definite water system. Their study revealed that the indices gave a better assessment of pollution levels.

Several statistical methods and models have been used for the assessment of groundwater quality and quantity in the world. For example, multivariate statistical techniques help to identify of possible factors/sources that influence water systems and offers a robust tool for reliable water resources management as well as quick solution to pollution problems in many part of the world including Bangladesh (Halim et al., 2010; Bhuiyan et al., 2011; Rahman et al., 2012a,b; Bhuiyan et al., 2015; Molla et al., 2015). Consequently, groundwater quality parameters that result in large data sets require to interpret complex data matrices, better understand the water quality and interrelationships between parameters and sampling sites. Therefore, statistical tools including principal components analysis (PCA) and cluster analysis (CA) coupled with Pearson correlation matrix analysis (CM) are used to resolve this complex data sets interpretation. On the other hand, geostatistical modeling is considered for spatial variation of water quality indices. The spatial distribution of contaminants in the groundwater reveals heterogeneity (Masoud, 2014). A various geostatistical interpolation method has been reported in different literature (Isaaks and Srivastava, 1989; Goovaerts, 1997; Webster and Oliver, 2001). In this study, ordinary kriging interpolation method is used for taking initial decision of spatial distribution of groundwater quality parameters. Thus, the cross validation results represent that the ordinary kriging technique is able to predict spatial variability more accurately for the study area. However, the objective of this paper is to develop a reliable multi-statistical method to characterize water quality of groundwater samples in Faridpur district of the central Bangladesh, which will be useful for decision makers to take proper initiative/s for groundwater quality management.

2. Materials and methods

2.1. Study area

Faridpur is located in central Bangladesh under Dhaka division. Geographically, it is positioned at 22.50–23.55° N and 89.15–90.40° E. The total area of this district is 2072.72 km^2 and the district is bounded by the Padma River to the north and east; across the river is in the central Bangladesh (Fig. 2). Groundwater is the main source of drinking and irrigation water in Faridpur district. Therefore, local people in the study area are fully depended on this groundwater for their daily life.

2.2. Sample collection and analytical procedure

Groundwater samples were collected from 60 preselected sampling points in the Faridpur district of central part of Bangladesh (Fig. 2). The geographical location of each pumping well was determined with a handheld global positioning system (GPS) (Explorist 200, Megellan). However, the sampling locations (S1 to S60) are shown in Fig. 2. The groundwater samples were collected randomly from the selected pumping wells, whose depths were varied from 14 to 204 m. Each well was pumped until steady pH and electrical conductivity were obtained. All water samples were collected in prewashed high-density polypropylene (HDPP) bottles following the standard method (APHA-AWWA-WEF, 2005). From each location, two sets of samples were collected by pre-washed plastic bottles. The sample bottles were kept in a cooler box and shifted to laboratory. The water samples for elemental analysis were acidified with HNO_3. Conversely, the samples collected for bicarbonate and major ions analysis were not acidified. All water samples were labeled and then kept frozen until chemical analysis.

All pH measurements were made using an Accumet electrode and Accumet Excel, XL50 (Dual channel pH/ion/conductivity) meter (Fisher Scientific, Singapore). The pH meter was standardized daily using a three-point calibration with pH 4 (SB101-500), pH 7 (SB107-500), and pH 10 (SB115-500) buffer solutions (Fisher Scientific, USA) before taking reading in samples. Conductivity meter was calibrated with reference solution just prior to measure electrical conductivity (EC). The concentrations of major anions (Cl^-, $HCO_3{}^-$ and $SO_4{}^{2-}$) concentration in water samples were measured by an ion chromatograph (761 Compact IC, Metrohm). The elemental concentration (i.e., Na, K, Ca, Mg, As, Fe, Mn, Ni, Pb and Zn) in water samples were measured by inductively coupled plasma mass spectrometry (Thermo Scientific X-Series2 ICP-MS), which was linearly calibrated from 10 to 100 μg/L with custom multi-element standards (SPEX CertiPrep, Inc., NJ, USA) before running the real samples. All solutions were prepared using double distilled deionized water. The accuracy and precision of analyses were tested through running duplicate

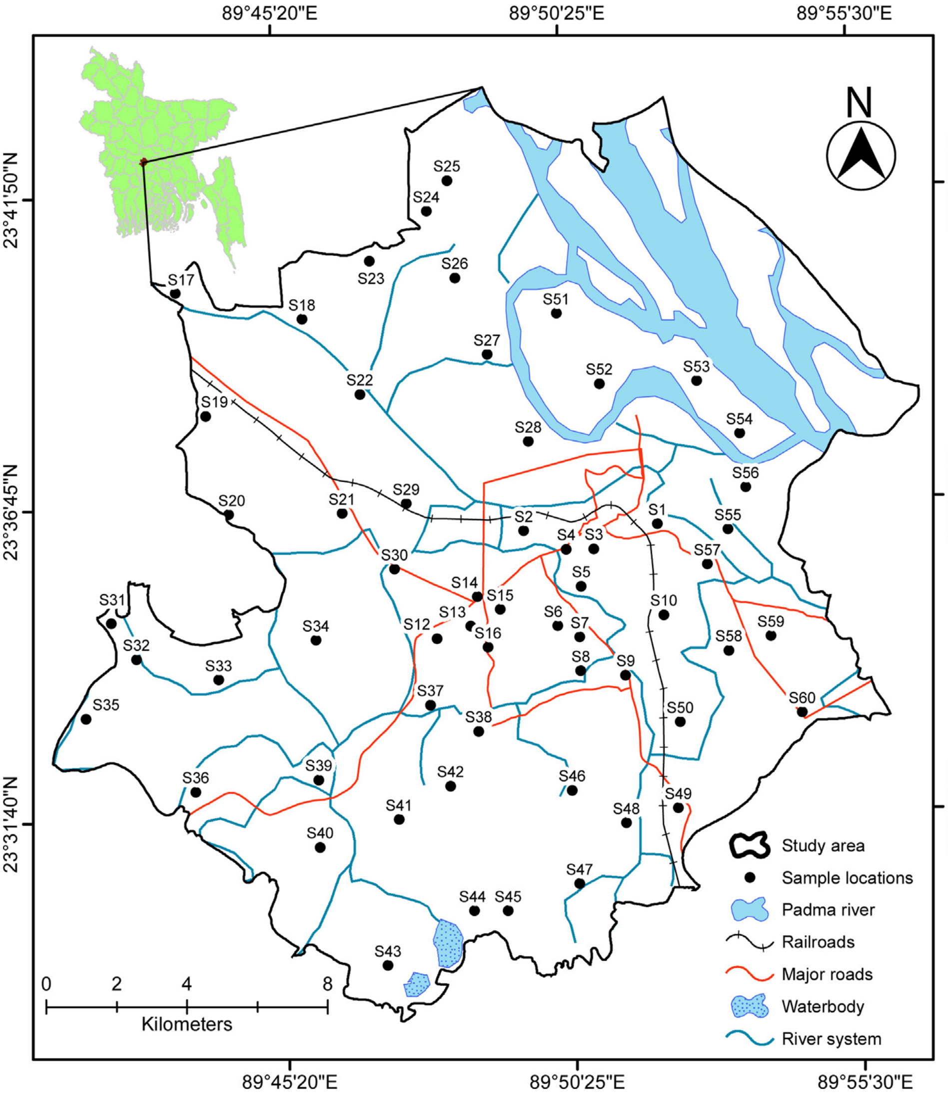

Fig. 2. Location map showing sampling site of the study area.

analyses on selected samples. Samples were diluted several times if needed, and the relative standard deviation of measured major ions was found to be within ±3%. The average results for all analyses were used to represent the data.

2.3. Drinking water evaluation indices

2.3.1. Groundwater quality index (GWQI)

Groundwater quality index (GWQI) method reflects the composite influence of the different water quality parameters on the suitability for drinking purposes (Sahu and Sikdar, 2008). The groundwater quality was measured by using the following equation for GWQI Vasanthavigar et al. (2010) with the respect to WHO and Bangladesh standards.

$$\mathrm{GWQI} = \sum \mathrm{SI}_i = \sum (W_i \times q_i) = \sum \left[\left(\frac{w_i}{\sum_n^{i=1} w_i} \right) \times \left(\frac{C_i}{S_i} \times 100 \right) \right] \tag{1}$$

Table 1
Parameters, weight factors, and limit values considered for the water quality index according to Nabizadeh et al. (2013) and Vasanthavigar et al. (2010).

Parameters	Units	Weight (w_i)	Relative weight (W_i)	Limit values
pH		4	0.09	6.5–8.5
Na^+	mg/l	4	0.09	200
K^+	mg/l	2	0.04	12
Ca^{2+}	mg/l	2	0.04	75
Mg^{2+}	mg/l	2	0.04	30
As	μg/l	4	0.09	50
Fe	μg/l	4	0.09	1000
Mn	μg/l	4	0.09	300
Pb	μg/l	4	0.09	10
Zn	μg/l	3	0.07	5000
Cl^-	μg/l	3	0.07	250
F^-	mg/l	4	0.09	1.5
HCO_3^-	mg/l	1	0.02	600
SO_4^{2-}	mg/l	4	0.09	400
		$\sum w_i = 45$	$\sum W_i = 1$	

where C_i is concentrations of each parameters, S_i is limit values, w_i is assigned weight according to its relative importance in the overall quality of water for drinking purposes (Table 1), q_i is water quality rating, W_i is the relative weight, and SI_i is the sub index of ith parameters.

2.3.2. Heavy metal pollution index (HPI)

The heavy metal pollution index (HPI) method has been established by assigning the weightage (W_i) for selected parameter and selecting the groundwater parameter on which the index has to be based. The rating is nearly zero to one, and its selection reveals the significance of each water quality parameter. It can be defined as inversely proportional to the recommended standard (S_i) for each parameter (Mohan et al., 1996). The concentration limits (i.e., the highest permissible value for drinking water (S_i) and maximum desirable value (I_i) for each parameter) were taken from the Indian Standard (2012) for this study. Heavy metal pollution index (HPI) was used for assigning a rating or weightage (W_i) for each selected parameter, is computed using the following equation (Mohan et al., 1996).

$$\mathrm{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \tag{2}$$

where, W_i is the sub-index of the ith parameter and W_i is unit weight of the ith parameter and n is the number of parameters. The sub-index Q_i is computed by

$$Q_i = \sum_{i=1}^{n} \frac{\{M_i(-)I_i\}}{(S_i - I_i)} \times 100 \tag{3}$$

where, M_i, I_i, and S_i donate for the 'monitored value', 'ideal value' and 'standard values' of the ith parameter respectively. The negative sign (−) denotes for numerical difference of the two values, ignoring the algebraic sign.

2.3.3. Heavy metal evaluation index (HEI)

Heavy metal evaluation index (HEI) method is consistent with the HPI method, which gives an insight the overall quality of the groundwater with respect to heavy metals (Prasad and Jaiprakas, 1999; Edet and Offiong, 2002), and it was calculated by the following equation:

$$\mathrm{HEI} = \sum_{i=1}^{n} \frac{H_c}{H_{mac}} \tag{4}$$

where, H_c is the monitored value and H_{mac} is the maximum admissible concentration (MAC) of ith parameters.

2.3.4. The degree of contamination (C_d)

The degree of contamination (C_d) is adopted from Backman et al. (1997), and the C_d was determined by the following equation:

$$C_\mathrm{d} = \sum_{i=1}^{n} C_{fi} \tag{5}$$

where, $C_{fi} = (C_{ai}/C_{ni}) - 1$ and C_{fi} is the contamination factor, C_{ai} is the analytical value and C_{ni} is the upper permissible concentration for the ith component, and n is indicated for the normative value. Here, C_{ni} is taken as maximum admissible concentration (MAC).

2.4. Multivariate statistical analysis

To evaluate the analytical data for finding source of pollutants, multivariate statistical techniques, e.g., correlation analysis, principal component analysis (PCA) and cluster analysis (CA) are commonly used in environmental studies (Mendiguchıía et al., 2004; Han et al., 2006; Rahman et al., 2014b). In our study, multivariate analysis was performed by using SPSS 22 for Windows.

PCA is widely used to reduce data and to extract a small number of latent factors for analyzing relationships among the observed variables (Farnham et al., 2003; Gou et al., 2007). PCA was performed to extract principal components (PC) from groundwater data and from all the sampling point, to evaluate spatial variations and possible source of heavy metals in groundwater. On the other hand, cluster analysis (CA) was performed to further classify elements of different sources on the basis of the similarities of their chemical properties (Rahman et al., 2014b). Hierarchical agglomerative clustering is the most common approach, which provides intuitive similarity relationships between any one sample and the entire data set, and is typically illustrated by a dendrogram (McKenna, 2003). The dendrogram provides a visual summary of the clustering processes, presenting a picture of the groups and their proximity, with a dramatic reduction in dimensionality of the original data. Cluster analysis was applied on experimental data standardized through z-scale transformation in order to avoid misclassification due to wide differences in data dimensionality (Liu et al., 2003).

The correlation coefficient matrix measures how well the variance of each constituent can be explained by relationships with each other (Liu et al., 2003). The terms “strong”, “moderate”, and “weak” were applied to factor loadings and refer to absolute loading values as >0.75, 0.75–0.50 and 0.50–0.30, respectively, following the approach of Liu et al. (2003).

2.5. Geostatistical modeling

We used ordinary kriging (OK) and semivariogram models for spatial distribution of the groundwater parameters, which related to groundwater application in hydrological studies. These interpolation techniques are well documented in the recent literature; (e.g. Masoud, 2014; Tapoglou et al., 2014). Kriging is one of the most popular and robust interpolation techniques among other techniques. It integrates both the spatial correlation and the dependence in the prediction of a known variable. Estimations of nearly all spatial interpolation methods can be represented as weighted averages of the sampled data. The following equation can be used to calculate spatial distribution (Delhomme, 1978):

$$\hat{z}(x_0) = \sum_{i=1}^{n} \lambda_i z(x_i) \tag{6}$$

where $\hat{z}$ is the estimated value of an attribute at the point of interest x_0, z is the observed value at the sampled point x_i, λ_i is the weight assigned to the sampled point, and n represent the number of sampled points used for the estimation (Webster and Oliver, 2001). The attribute is usually called the primary variable, especially in geostatistics. The semivariance can be estimated from the groundwater data by the following equation:

$$\gamma(h) = \frac{1}{2n} \sum_{i=1}^{n} [z(x_i) - z(x_i + h)]^2 \tag{7}$$

where, n is the number of pairs of sample points separated by standard distance called lag h (Burrough and McDonnell, 1998). The $z(x_i)$ is the value of the variable z at location of x_i. Variogram modeling and estimation are important for structural analysis and spatial interpolation. Ordinary kriging use the above equations to estimate the prediction of groundwater data. Ordinary Kriging estimates the local constant mean (Goovaerts, 1997). Out of different kriging techniques, the ordinary kriging (OK) method is used in the present study because of its simplicity and prediction accuracy in comparison to other Kriging methods (Gorai and Kumar, 2013). Different semivariogram models from the more common like the linear, the exponential and the spherical have been tested and validated all over the world in recent past (Varouchakis and Hristopulos, 2013). These semivariogram models were chosen because they show various types of semivariogram models; the exponential is a differentiable variogram, spherical is best fitted variogram, the Gaussian is a non-differentiable and the linear signify the simple one. The best-fitted theoretical semivariograms models for all groundwater parameters were prepared based on selecting the trial and error parameters. Predictive performances of the fitted models were checked on the basis of cross validation tests (Gorai and Kumar, 2013). The mean error (ME), mean square error (MSE), root mean error (RMSE), average standard error (ASR) and root mean square standardized error (RMSSE) values were assessed to establish the fitted models performances. Models attained the best goodness of fit resulted in minimum mean error (ME), root mean error (RME), mean squared error (MSE), attained root mean squared error (RMSE) and average squared error (ASE) close to unity are considered the best fit models performances (ESRI, 2009). RMSE is used to investigate the best model by comparing its value, and the smallest value of RMSE indicates the most suitable model to the data (Marko et al., 2014). After completing the cross validation process, kriging maps were produced which give insight a graphical representation of the distribution of the groundwater quality parameters. These kriged maps were generated by the Arc GIS (10.2 version).

3. Results and discussion

3.1. General characteristics of groundwater quality

General characteristics of groundwater physicochemical parameters for the study area are summarized in Table 2 with minimum, maximum, mean, and standard deviation values. All the samples ($n = 60$) showed the pH values ranged from 6.38 and 7.35 with a mean value of 6.91 ± 0.19, indicating acidic to slight alkaline in nature. The findings of Rahman et al. (2014a) are consistent with this study, which assessed the quality of groundwater in the

Table 2
Descriptive statistics of physiochemical parameters and heavy metal in the study area.

Parameters	Minimum	Maximum	Mean	Std. deviation	Variance	Standards			
						WHO (2011)	BMAC (1997)	Indian standard (2012)	
								Acceptable limit	Permissible limit
pH	6.38	7.35	6.9122	0.19034	0.036	–	6.5–8.5	6.5–8.5	–
EC (μs/cm)	344	1760	788.7667	242.8307	58 966.72	–	1000	–	–
Na^+ (mg/l)	5.1	175	35.3467	37.9988	1443.909	–	200	–	–
K^+ (mg/l)	2.6	8.6	5.0083	1.3651	1.863	–	12	–	–
Ca^{2+} (mg/l)	34.8	190	103.7533	36.23135	1312.711	–	75	75	200
Mg^{2+} (mg/l)	8.92	59.5	32.9153	10.38636	107.877	–	30–35	30	100
As (μg/l)	8	1460	118.6	195.525	38 230.01	10	50	10	50
Fe (μg/l)	52	19 600	5951.6	4931.166	24 316 403	–	300–1000	300	–
Mn (μg/l)	0.04	4.23	0.6388	0.73469	0.54	–	100	100	300
Ni (μg/l)	1.1	18.8	3.2817	2.33793	5.466	70	100	20	–
Pb (μg/l)	0.02	28.6	0.6272	3.67516	13.507	10	50	10	–
Zn (μg/l)	2	58	10.3833	10.19985	104.037	–	5000	5000	15 000
Cl^- (mg/l)	1.8	195	23.955	36.64463	1342.829	250	150–600	250	1000
F^- (mg/l)	0.02	0.4	0.1213	0.06182	0.004	1.5	–	1	1.5
HCO_3^- (mg/l)	200	848	542.3667	130.7132	17 085.93	–	600[a]	–	–
SO_4^{2-} (mg/l)	1.1	12.3	5.2665	2.4024	5.772	–	400	200	400

BMAC (Bangladesh maximum admissible concentration).

northwestern part of Bangladesh and found to be acidic to little alkaline water. The EC in the study area varied from 344 to 1760 μs/cm with a mean value of 788.76 ± 242.83 μs/cm. Among the cations in studied samples, the concentrations of Na^+, K^+, Ca^{2+}, and Mg^{2+} ions ranged from 5.1 to 175 mg/l, 2.6 to 8.6 mg/l, 34.8 to 190 m/l, and 8.92 to 59.5 mg/l with the mean values of 35.34 ± 37.99 mg/l, 5.00 ± 1.36 mg/l, 103.75 ± 36.23 mg/l, and 32.91 ± 10.38 mg/l, respectively. The concentration of dissolved anions like Cl^-, F^-, HCO_3^- and SO_4^{2-} varied from 1.8 to 195 mg/l, 0.02 to 0.4 mg/l, 200 to 848 mg/l, and 1.1 to 12.3 mg/l with the mean concentrations of 23.95 ± 36.64 mg/l, 0.12 ± 0.06 mg/l, 542.36 ± 130.71 mg/l and 5.26 ± 2.40 mg/l, respectively. The chronological order of major cations of the groundwater samples are $Ca^{2+} > Na^+ > Mg^{2+} > K^+$, and major anions are $HCO_3^- > Cl^- > SO_4^{2-} > F^-$. Quddus and Zaman (1996) noted that major ions present in groundwater are Ca^{2+}, Mg^{2+}, HCO_3^-, Na^+, Cl^- and SO_4^{2-} in the western part of Bangladesh. Moreover, highest concentration of HCO_3^- and Ca^{2+} ions revealed that the study area might be influenced by carbonate mineral dissolution. Holland (1978) pointed out that 74 ± 10% of Ca^{2+} and 40 ± 20% of Mg^{2+} in groundwater derived from dissolution of carbonate minerals rather than come from silicate minerals. The semi metal and heavy metals like As, Fe, Mn, Ni, Pb, and Zn concentrations were found to be ranged from 8 to 1460 μg/l, 52 to 19 600 μg/l, 0.04 to 4.23 μg/l, 1.1 to 18.8 μg/l, 0.02 to 28.6 μm/l and 2 to 58 μg/l, respectively. The mean concentration of heavy metals is followed the descending order: Fe > As > Zn > Ni > Mn > Pb. However, the mean value of Fe (5951.6 ± 4931.16 μg/l) and As (118.6 ± 195.52 μg/l) are higher than the water quality standards set by Bangladesh (Bangladesh Standard, 1997) and international organization (WHO, 2011). It is found that most of the groundwater samples showed the high concentrations of Fe, As and Zn values in the study area. Noteworthy, the high concentration of total As is associated with Fe and Zn in shallow groundwater systems. Reza et al. (2010) reported that high As, low Fe, low Mn found in groundwater of Meghna flood plain of southeastern part of Bangladesh but this finding differed from the observation in this lower Padma River system of study area where very high concentration of Fe, Mn and total As.

3.2. *Evaluation of drinking water quality*

Ground water quality index (GWQI) is defined as a technique of rating that provides the composite influence of individual water quality parameters on the overall quality of water for human consumption. To determine suitability of the groundwater quality for drinking water purposes using international standard (WHO, 2011) and Bangladesh standard (1997) values following Eq. (1) and the results are presented in Table 3. The GWQI values ranged from 27.59 to 326 with a mean value of 100.7. Results revealed that about 48% of the samples (S10, S13, S15, S17, S19, S20, S21, S23, S24–S26, S27, S28, S29, S30, S32, S34, S41, S48–S50, S51, S53–S56, S57 S60) fall below the critical limit of GWQI. Furthermore, in case of GWQI, 28.33% samples (S19, S20, S21, S23, S24–S26, S28, S29, S30, S34, S51, S53–S56, S60) reported excellent water quality type, and 18.33% (S10, S13, S15, S17, S27, S32, S41, S48–S50, S57) represented good water quality type, whereas 48.33% (S2–S7, S9, S11, S12, S14, S16, S18, S22, S31, S33, S35–S37, S39, S40, S42–47, S52, S58, S59) exhibited poor water quality, 3.33% (S8, S38) of the water was of very poor quality type and rest of 1.67% (S1) indicated unsuitable water for drinking water uses (Table 4).

On the other hand, the degree of contamination (C_d) was used as a reference of estimating the level of pollution. This study revealed that the ranges and mean value of C_d of the groundwater samples were 0.19–32.63 and 7.51 respectively (Table 3). The computed C_d values were found to be in exceeding 3, indicating 73.33% of the samples exceeded the critical values considering the classification of C_d values (Edet and Offiong, 2002). Therefore, C_d values have been suggested that most of the samples locations are highly polluted in the study area. Fe, Mn, Pb, Zn, Ni and As were used to calculate heavy metal pollution index (HPI) and heavy metal evaluation index (HEI) using the Indian standard (2012) values of metal contents in groundwater samples. This study shows that the range and mean value of HPI for the groundwater samples are 5.96–425.89 and 45.09 respectively (Table 3). But interestingly, same sample locations showed less polluted in most of the area in term of HPI values compare to C_d values. The critical value for HPI is 100, indicating 6.67% of the samples fall above the critical values. However, the HEI values ranged from 0.89 to 33.65 with a mean value of 8.55; indicating all sample locations fall below the critical values (Table 3). Different HEI criteria values have been developed for groundwater, depending on their respective mean values, and the different levels of contamination were demarcated by a multiple of the mean values. The proposed HEI criteria for the ground water samples were thus classified: low (HEI < 45), medium (HEI = 45–90) and high (HEI > 90). The HEI was used for a better understanding of the pollution indices developed by Edet and Offiong (2002).

Table 3
Groundwater indices evaluation.

Drinking water quality indices				
Sample ID	GWQI	C_d	HEI	HPI
S1	326	32.63	33.65	425.89
S2	166.34	14.6	15.9	98.58
S3	104.91	6.39	8.32	194.99
S4	100.78	7.58	8.27	17.11
S5	108.84	7.72	9.8	55.89
S6	135.22	11.08	12.65	32.88
S7	192.33	17.87	19.23	32.37
S8	208.3	19.22	20.72	32.26
S9	101.38	7.4	8.52	18.03
S10	82.33	5.27	7.35	59.02
S11	110.67	8.38	8.98	14
S12	187.47	16.69	18.41	46.94
S13	89.33	7.58	6.36	15.78
S14	127.29	9.72	11.61	54.58
S15	64.88	2.88	3.08	20.19
S16	129.91	10.36	11.11	19.25
S17	96.81	6.6	8.28	44.7
S18	121.98	9.64	11.08	78.9
S19	33.14	0.43	1.62	21.92
S20	41.92	2.52	1.14	9.98
S21	38.34	0.44	1.99	17.98
S22	137.91	11.18	13.01	52.23
S23	39.32	1.54	2.68	37.9
S24	33.91	0.39	1.73	23.05
S25	37.33	1.65	1.29	18.69
S26	40.47	1.82	1.38	15.32
S27	64.28	3.19	4.72	26.21
S28	35.13	0.35	1.65	12.49
S29	29.31	0.23	0.96	6.81
S30	33.05	0.19	1.53	15.36
S31	125.97	10.05	11.85	47.64
S32	97.72	6.96	8.38	45.8
S33	124.52	9.8	11.31	46.6
S34	27.59	0.19	0.89	5.96
S35	135.02	11.45	12.26	20.64
S36	108.11	7.98	9.8	29.86
S37	141.52	11.87	13.16	35.64
S38	210.66	19.37	19.73	30.9
S39	133.09	10.66	12.46	104.47
S40	109.19	7.89	9.99	38.55
S41	81.37	5.22	6.66	18.71
S42	121.45	9.38	10.94	62.33
S43	153.73	12.73	14.68	46.64
S44	158.34	13.64	15.06	90.26
S45	100.77	7.04	8.63	33.3
S46	100.28	6.52	8.27	51.32
S47	104.81	7.11	8.69	59.17
S48	98.35	6.47	7.78	27.87
S49	80.75	4.84	6.71	50.42
S50	99.73	6.79	8.51	18.31
S51	35.02	0.74	1.43	20.07
S52	104.53	7.92	8.93	35.47
S53	47.59	3.16	1.8	25.68
S54	44.53	3.21	1.45	18.42
S55	38.85	2.01	1.37	12.18
S56	40.19	1.78	1.57	9.59

Table 3 (*Continued*)

Drinking water quality indices				
Sample ID	GWQI	C_d	HEI	HPI
S57	54.36	2.1	3.64	24.2
S58	175.21	15.46	16.95	123.93
S59	123.74	11.36	10.45	15.02
S60	46.39	1.94	2.84	37.58
Minimum	27.59	0.19	0.89	5.96
Maximum	326	32.63	33.65	425.89
Mean	100.704	7.5197	8.554	45.097
Critical value (CV)	100	>3		100
Percentage of samples exceedingCV	53.33%	73.33%		6.67%

A comparison between C_d, HEI and HPI values in groundwater quality schemes are presented in Table 4. The C_d values indicated low level of pollution; with 71% of the samples (S3–S5, S9–S11, S13–S15, S17–S21, S23–S30, S32–S34, S36, S40–S42, S45–S57, and S60) while 26.67% (S2, S6–S8, S12, S16, S22, S31, S35, S37–S39, S43, S44, S58, and S59) had medium levels of pollution fall below the mean value. Similar observation made for HPI and HEI values were also low with 66.67% (S4, S6–S9, S11, S13, S15–S17, S19–S21, S23–S30, S34–S38, S40, S41, S45, S48, S50–S57, S59, and S60) and 65% (S3–S5, S9–S11, S13, S15, S17, S19–S21, S23–S30, S32, S34, S36, S40, S41, S45–S57, and S60) of the samples fall below the mean value. On the other hand, a significant correlation was observed among water quality indices (GQWI, HPI, HEI, and C_d) values, heavy metal (Fe, Zn, Mn, Ni, Pb, and Zn), and toxic heavy metal (As) concentrations. The As and Fe showed most significant correlation with all water evaluation indices.

Table 4
Classification of the ground water quality of the study area based on modified categories of quality indices values.

Index method	Category	Degree of pollution/Water class	Number of locations	% of sample	Samples
HPI	<45	Low	40	66.67	S4, S6–S9, S11, S13, S15–S17, S19–S21, S23–S30, S34–S38, S40, S41, S45, S48, S50–S57, S59, S60
	45–90	Medium	15	25	S5, S10, S12, S14, S18, S22, S31–S33, S42–S44, S46, S47, S49
	>90	High	5	8.33	S1–S3, S39, S58
HEI	<10	Low	39	65	S3–S5, S9–S11, S13, S15, S17, S19–S21, S23–S30, S32, S34, S36, S40, S41, S45–S57, S60
	10–20	Medium	19	31.67	S2, S6, S7, S12, S14, S16, S18, S22, S31, S33, S35, S37–S39, S42–S44, S58, S59
	>20	High	2	3.33	S1, S8
C_d	<10	Low	43	71.66	S3–S5, S9–S11, S13–S15, S17–S21, S23–S30, S32–S34, S36, S40–S42, S45–S57, S60
	10–20	Medium	16	26.67	S2, S6–S8, S12, S16, S22, S31, S35, S37–S39, S43, S44, S58, S59
	>20	High	1	1.67	S1
GWQI	<50	Excellent water	17	28.33	S19, S20, S21, S23, S24–S26, S28, S29, S30, S34, S51, S53–S56, S60
	50–100	Good water	11	18.33	S10, S13, S15, S17, S27, S32, S41, S48–S50, S57
	100.1–200	Poor water	29	48.33	S2–S7, S9, S11, S12, S14, S16, S18, S22, S31, S33, S35–S37, S39, S40, S42–47, S52, S58, S59
	200.1–300	Very poor water	2	3.33	S8, S38
	>300	Water unsuitable for drinking purposes	1	1.67	S1

Table 5
Correlation coefficient matrix for indices values and metal concentration.

	HPI	HEI	C_d	GWQI
HPI	1	**.640**[**]	**.634**[**]	**.637**[**]
HEI	**.640**[**]	1	**.989**[**]	**.997**[**]
C_d	**.634**[**]	**.989**[**]	1	**.994**[**]
GWQI	**.637**[**]	**.997**[**]	**.994**[**]	1
As	**.933**[**]	**.634**[**]	**.632**[**]	**.624**[**]
Fe	0.059	**.786**[**]	**.775**[**]	**.789**[**]
Mn	0.035	−0.213	−0.178	−0.208
Ni	−0.01	−0.172	−0.129	−0.16
Pb	.328[*]	−0.004	−0.023	0.01
Zn	−0.031	−0.151	−0.147	−0.159

Bold digits are significant at 95% and 99% confidence level as denoted by * (95%) and ** (99%).
[*] Correlation is significant at the 0.05 level (2-tailed).
[**] Correlation is significant at the 0.01 level (2-tailed).

It has been suggested that As and Fe are major contributory parameters to all indices (Table 5). Hence, comparison of three classification schemes; the values of C_d, HEI and HPI are falling below their respective mean values indicate low level of pollution. However, C_d values are good agreement with GWQI indices values. Bhuiyan et al. (2015) noted that HEI gave better assessment result for heavy metal pollution in Dhaka, Bangladesh.

3.3. Source of pollution and factor controlling of groundwater quality

Principal component analysis (PCA) was performed on the groundwater quality data using Varimax rotation with Kaiser Normalization, which was used to elucidate the observed relationship of cluster variables in simple ways, expressed in the patterns of variance and covariance between the variable and similarities between observations. This technique exhibits a complex hydrochemistry in the area along with interplay of ions exchange, leaching of material, agricultural fertilizer, domestic sewage and weathering of minerals. Kaiser proposed the use of only factors with eigenvalues exceeding one (Liu et al., 2003). Six factors were extracted for groundwater quality data sets based on eigenvalues more than 1, which represented 80.56% of total variance in the study area (Fig. 3(a) and (b)). On the other hand, the scree plot (Fig. 3(a)) was used to identify the number of PCs to be retained to understand the underlying parameters structure. The calculated factor loadings, together with cumulative percentage, and percentages of variance explained by each factor are shown in Table 6. About 52.26% of the total variance was represented in the first three

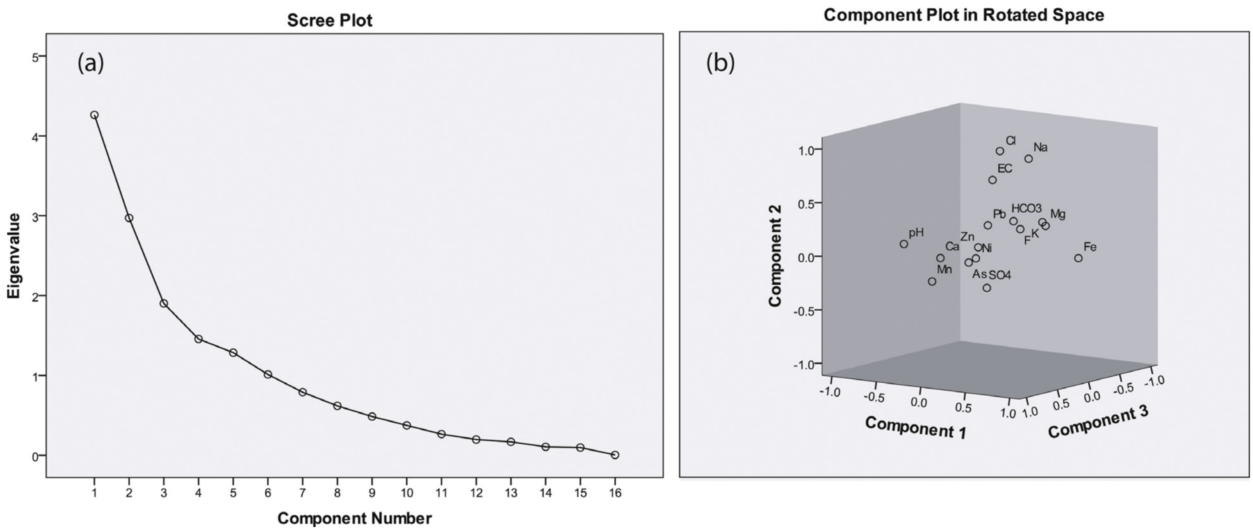

Fig. 3. Principal component analysis by (a) scree plot of the characteristic roots (eigenvalues), and (b) component plot in rotated space.

Table 6
Varimax rotated principal component analysis for groundwater samples.

Parameters	PC1	PC2	PC3	PC4	PC5	PC6
R mode						
pH	−0.885	−0.007	0.117	0.096	0.108	−0.063
EC	0.333	0.760	0.433	0.132	0.024	−0.056
Na^{+}	0.254	0.850	−0.260	−0.080	0.099	0.107
K^{+}	0.655	0.349	0.090	−0.122	−0.178	−0.143
Ca^{2+}	0.071	0.071	0.896	0.292	−0.054	−0.003
Mg^{2+}	0.776	0.344	0.213	0.098	−0.012	0.053
As	−0.135	−0.100	0.147	−0.095	0.915	−0.076
Fe	0.686	−0.061	−0.444	−0.072	0.222	−0.062
Mn	−0.090	−0.178	0.799	−0.073	−0.068	0.028
Ni	0.121	0.001	0.397	0.812	−0.029	0.053
Pb	−0.036	0.233	−0.019	0.081	0.085	0.854
Zn	−0.179	0.007	−0.069	0.905	−0.043	−0.058
Cl^{-}	0.064	0.930	−0.071	0.005	−0.050	0.070
F^{-}	0.065	0.154	−0.396	0.018	0.802	0.066
HCO_3^{-}	0.634	0.419	0.526	0.204	0.064	0.030
SO_4^{2-}	0.036	−0.327	0.100	−0.241	−0.310	0.527
Eigenvalues	4.262	2.972	1.902	1.457	1.285	1.012
% of variance	18.449	17.599	16.220	10.926	10.650	6.721
Cumulative %	18.449	36.047	52.267	63.193	73.843	80.564
Sites Q mode						
S1	−0.806	−0.093	1.596	0.012	6.508	−0.624
S2	−0.040	−0.349	0.336	0.106	1.160	0.621
S3	−0.288	1.823	−0.101	0.574	0.692	6.472
S4	−0.085	0.714	−0.285	0.701	0.282	−0.906
S5	0.083	−0.427	−1.204	0.200	0.148	−0.255
S6	1.403	−1.129	−0.911	−0.317	−0.106	0.620
S7	2.138	−1.594	−1.637	1.639	−0.202	0.973
S8	2.315	−0.992	−1.401	−0.469	−0.220	−0.258
S9	0.601	0.015	−0.660	0.005	0.698	−0.134
S10	−1.177	−1.045	−1.267	−0.855	−0.151	1.372
S11	0.328	1.675	−0.634	−0.313	−0.259	−0.494
S12	1.827	−0.468	−1.168	0.332	0.491	−0.584
S13	−0.691	2.872	0.358	0.765	−0.410	−0.892
S14	0.424	−0.275	−0.732	−0.304	0.760	−0.060
S15	−0.678	3.754	−0.255	0.401	−0.212	−0.593
S16	0.957	1.245	−0.532	−0.687	−0.007	0.898
S17	−0.076	−0.621	−0.044	0.726	−0.028	0.127
S18	−0.400	−0.533	1.769	−0.597	0.049	−0.298
S19	−1.883	−0.212	−1.266	1.519	−0.863	−0.505
S20	0.001	0.842	1.576	−0.058	−0.581	0.901
S21	−1.246	−0.365	−0.384	1.236	−0.733	−0.589
S22	−0.022	−0.424	−0.334	−0.273	−0.080	−0.337
S23	−2.404	−0.566	−0.964	−0.966	−0.501	0.140
S24	−1.940	−0.231	−0.517	−0.380	−0.591	0.158
S25	0.110	−0.228	1.571	−0.007	−0.784	−0.142
S26	0.079	0.615	1.185	0.035	−0.896	−0.636
S27	−0.600	−0.137	0.610	−0.281	−0.277	−0.851
S28	−1.054	−0.419	0.234	−0.825	−0.966	0.264
S29	−1.079	−0.463	−0.181	−0.714	−0.594	0.894
S30	−1.174	−0.625	−0.014	−0.572	−0.553	0.190
S31	−0.473	−0.718	−0.545	0.041	0.361	−0.117
S32	−0.448	−0.527	0.440	−0.386	−0.238	0.628
S33	−0.053	−0.602	0.399	−0.468	−0.058	0.410
S34	−1.718	−0.216	−0.508	0.404	−0.428	−0.695
S35	0.446	−0.173	−0.016	0.119	0.006	0.061
S36	−0.583	−0.644	−0.518	0.062	0.092	0.057

Table 6 (*Continued*)

Parameters	PC1	PC2	PC3	PC4	PC5	PC6
S37	0.744	−0.328	−0.346	−0.128	0.306	−0.659
S38	1.580	2.776	−0.532	−0.009	−0.128	−0.772
S39	−0.541	−0.525	0.056	−0.633	0.570	0.021
S40	−0.372	−0.722	−0.712	−0.325	−0.075	0.023
S41	−0.546	−0.512	−0.020	0.010	0.057	−0.192
S42	0.138	−0.417	−0.141	−0.322	0.533	−0.203
S43	0.710	−0.402	−0.517	−0.571	0.003	−0.291
S44	0.300	−0.333	0.059	−0.004	0.736	−0.550
S45	0.559	−0.223	−0.188	0.009	−0.197	−0.521
S46	−0.005	0.746	−0.790	−0.258	0.538	−0.394
S47	−0.229	1.843	−0.795	−0.648	0.649	−0.376
S48	0.984	1.573	−1.158	−1.119	−0.235	−0.519
S49	−0.303	−0.259	−0.741	−0.711	−0.263	−0.745
S50	1.126	−0.066	−1.077	−0.516	0.048	0.115
S51	−0.815	−0.198	0.554	−0.601	−0.890	0.102
S52	0.134	−0.519	2.374	−1.027	0.059	0.039
S53	1.526	−0.255	2.502	−0.438	−0.915	0.341
S54	1.452	−0.322	2.669	−0.582	−1.046	0.612
S55	0.278	0.154	1.208	1.577	−0.784	−0.895
S56	−0.037	0.044	1.649	−0.062	−0.829	−0.560
S57	−0.643	−0.376	0.837	0.182	−0.093	−0.276
S58	0.309	−0.296	0.050	−0.342	0.924	−0.412
S59	1.893	−0.334	0.488	0.044	−0.527	0.108
S60	−0.038	−0.527	0.574	6.066	0.051	0.191

loading factors in the groundwater samples (Fig. 3(b)). The PC1, PC2, PC3, PC4, PC5, and PC6 for groundwater quality data were elucidated the total variance of 18.44%, 17.59%, 16.22%, 10.92%, 10.65%, and 6.72% respectively.

The study also revealed that PC1 in the data sets explained 18.44% of total variance, and it was positively loaded with K^+, Mg^{2+}, Fe, and $HCO_3{}^-$, which were mostly distributed in S6–S8, S12, S16, S38, S48, S50, S53–S54, and S59 sample locations, indicating geogenic hydrogeochemical evolution of groundwater by rock–water interaction with ions exchange (Omo-Irabor et al., 2008). PC1 showed strong loading of K^+ and Mg^{2+} are indicative of lithologic influence along with leaching of secondary salts by rain water. However, $HCO_3{}^-$ reflecting intensive weathering of carbonate environments, and alkaline nature of groundwater (Rahman et al., 2014a). Natural sources like oxidation of iron (Rahman and Gagnon, 2014) and rain water through the leaching of secondary salts infiltrated into aquifer may be an option for possible source of groundwater contamination. The PC2 in the data sets explained 17.59% of total variance, and it was positive loaded with EC and Na^+, which were significantly distributed in S3, S11, S13, S15, S16, S38, and S47–S48. The PC2 were also derived from mixed sources such as salinity from the sea water, use of chemical fertilizers in agricultural fields. Because the study area was not very far from the salinity affected areas of Padma River systems. This trend exhibited the leaching of secondary salt precipitated when anthropogenic activities occurred in the study area. The PC3, accounting for 16.22% of total variance, was loaded on Ca, Mn and $HCO_3{}^-$, which were widely distributed in S1, S18, S20, S25–S26, and S52–S56 respectively, and could be ascribed to geochemical alteration of carbonate minerals. The Ca^{2+} is abundant in rocks and soil, particularly carbonate bearing rock. $HCO_3{}^-$ may be caused by oxidation of organic waste in groundwater (Rahman et al., 2013). The Mn might be originated by geogenic sources, which could be released by chemical weathering of parent materials. PC4 had loaded with Ni and Zn, which were distributed for S7, S19–S20, S55, and S60 accounting for 10.92% of total variance related to anthropogenic sources. Potential heavy metals (Ni and Zn) can be assimilated in groundwater through leaching of metals from industrial activities, burning surrounding in the study area. Several industries such as ceramics, brick and pottery industries are located in the study area, which are responsible for heavy metal (Ni and Zn) pollution. The As and F^- were important parameters in S1–2 and S58, as PC5 retained high positive loading in these samples with 10.65% of total variance, indicating geogenic origin of groundwater pollution. Elevated As levels in precipitation and in soils surrounding smelters have frequently been documented in worldwide (Beaulieu and Savage, 2005). It should be noted here that the study area is also identified as As affected area (Zaman et al., 2001). The PC6 had low to moderate loadings of Pb and SO_4^{2-} for S3, S7, S10, S16, S20, and S29 samples with a 6.72% of total variance, showing the effect of agricultural and stagnant water (ponds and tank), fertilizers in the sample sites.

3.4. Spatial similarity and sampling sites grouping

The R-mode cluster analysis (CA) retained three main clusters for data sets of analyzed parameters. R-mode cluster analysis was applied to predict physicochemical groupings in the groundwater datasets, and the results are shown in Fig. 4(a). Parameters belonging to the same cluster were likely to be found from a same source. Cluster 1 included EC, Na^+, Cl^-, Mg^{2+}, K^+, HCO_3^-, Fe and Pb, which might be explained by combining mix sources, trace elements and leaching of fertilizers from the soil horizon to the aquifer. Cluster 2 consists of Mn, Zn, Ca, Ni and it reflected the influence of domestic and agricultural pollution (Omo-Irabor et al., 2008). Cluster 3 included As, SO_4^{2-}, pH, F^- elucidated by the dissolution of minerals under basic condition. The CA results mostly good agreed with that of PCA.

Q-mode CA was used to recognize the spatial similarities and site grouping among the sampling points. Particular group/class shows similar characteristics with respect to the analyzed parameters in a samples cluster. The 60 sampling

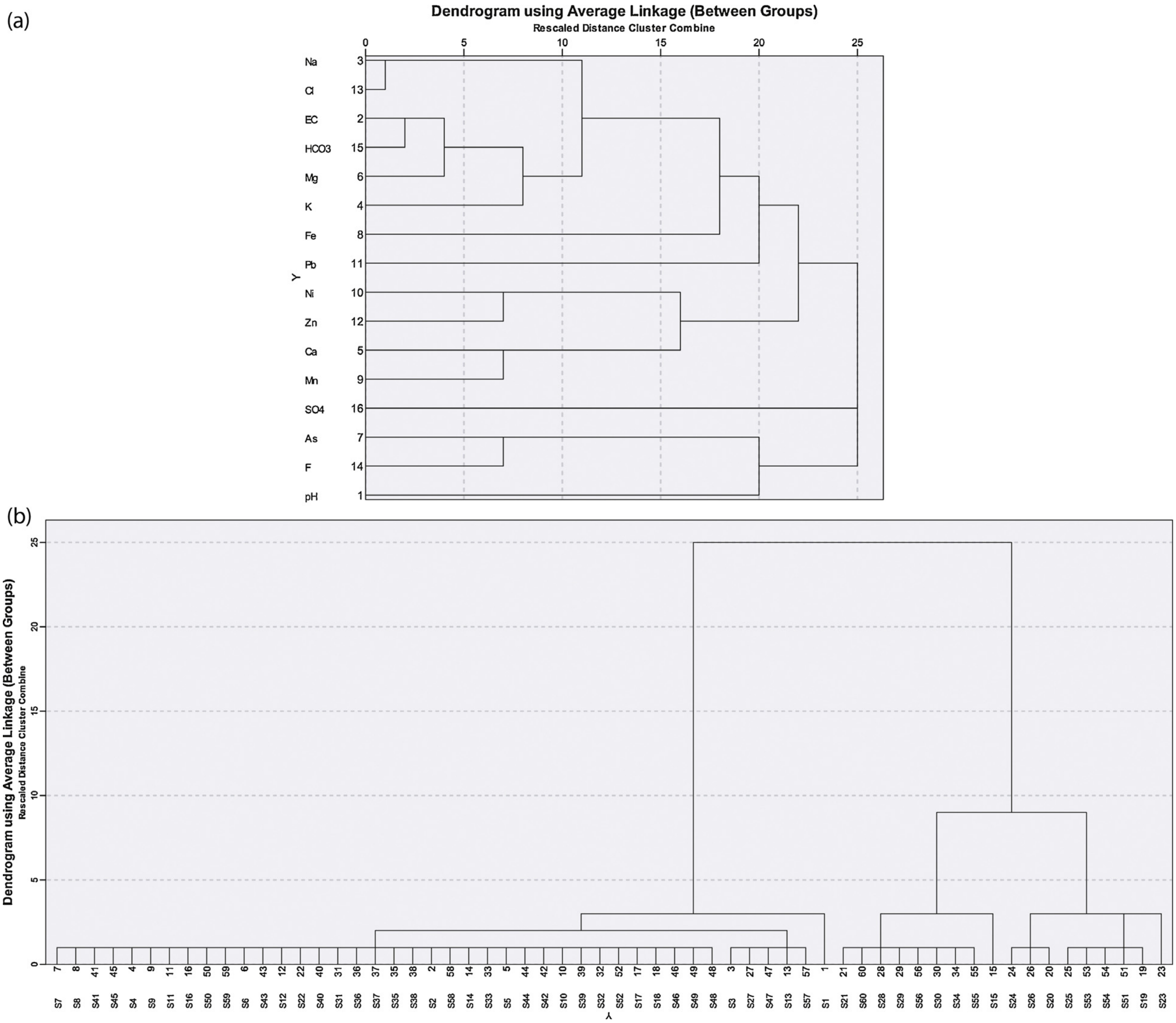

Fig. 4. (a) Dendrogram showing the hierarchical clusters of analyzed parameters. (b) Dendrogram showing the hierarchical clusters of analyzed samples site.

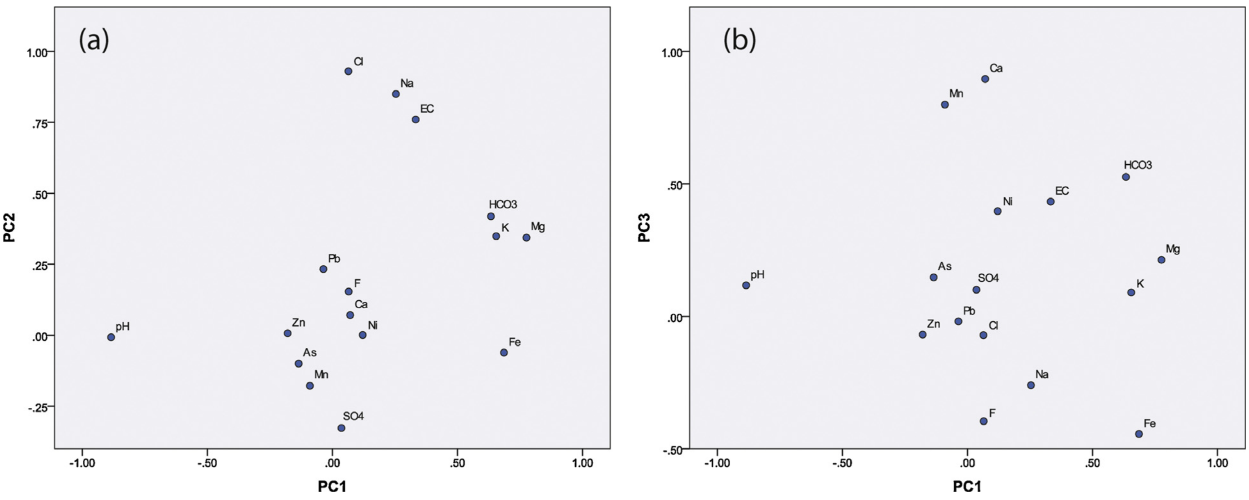

Fig. 5. Plots of first three principal component loadings, (a) PC1 vs. PC2 and (b) PC1 vs. PC3 for all analyzed parameters.

sites for ground water fall into three clusters (Fig. 4(b)). Cluster 1 consists of 42 sampling points. These 42 sampling points are S1–S14, S16–S18, S22, S27, S31–S33, S35–S50, S52, S57, S58, and S59. This cluster 1 site is closed to groundwater contamination via agricultural fertilizer. S1 and S39 lies at the distal part of the study area, they have different hydrogeochemistry from other others as indicated by its linkage distances. Likewise, S13 and S37 sites are occurring at the distal part of domestic sewage drainage channel connected the Pleistocene terrace (Madhupur clay formation) and retain water from this formation. Cluster 2 had the following 9 sample sites S15, S21, S28, S29, S30, S34, S55, S56, and S60, whereas clusters 3 consists of 9 sites which were S19, S20, S23–S26, S51, S53, and S54. These sites are mostly less contamination of groundwater due to leaching of parent material and agricultural runoff. It is thus not surprisingly that cluster 2 (S15 and S28), cluster 3 (S23 and S26) were grouped together in the similar clusters. The associations among the analyzed parameters were also visualized in the factor loadings plots of PC1 vs. PC2 and PC1 vs. PC3 (Fig. 5(a) and (b)). For all parameters, six main clusters were obtained from the plotting of PC1 vs. PC2 (Fig. 5(a)). Cluster 1 contained only pH. Cluster 2 consists of Pb, F^{-}, Ca, and Ni. Cluster 3 included Zn, As, Mn, SO_4^{2-}, Fe independently remained in cluster 4. Cluster 5 contains HCO_3^{-}, K^{+}, Mg^{2+} and cluster 6 belongs to Cl^{-}, Na^{+} and EC. Near similar groupings of parameters were observed on the plot of PC1 vs. PC3 (Fig. 5(b)). Cluster 1, 4 and 5 are similar in plotting of PC1 vs. PC2 and PC1 vs. PC3. It is found that plot of PC1 vs PC3 showed more variation in groundwater parameters than PC1 vs PC2. In Fig. 6 sample sites were plotted on the plane of the PC1 vs

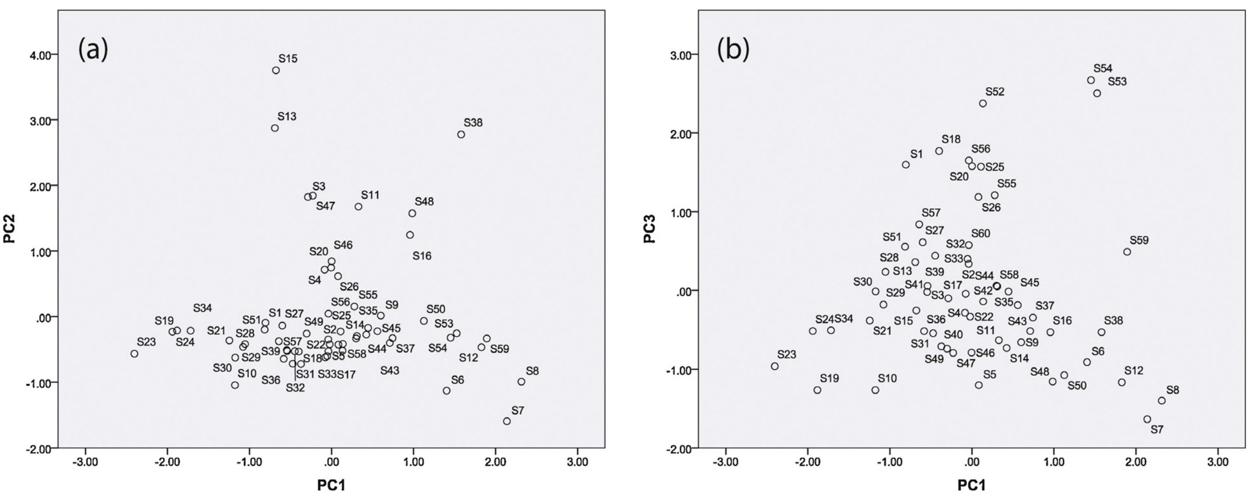

Fig. 6. Plots of first three principal component loadings, (a) PC1 vs. PC2, (b) PC1 vs. PC3 for all sampling site.

PC2 of Q-mode PCA, PC1 separated S3, S7–8, S11, S13, S15–16, S20 and S46–48 from the other sites (Fig. 6(a)). Similarly, the first two components (PC1 vs PC3) of Q-mode PCA, PC1 separated S1, S-7–8, S10, S18–20, S23, S50, S52–54 and S59 from other sites in the study area (Fig. 6(b)).

3.5. Correlation matrix analysis

The Pearson's correlation coefficient was used to show the interrelationship and coherence pattern among groundwater quality parameters. The correlation coefficient values of the analyzed water quality parameters are given in Table 7. Correlation matrix showed inter-parameter relationships agreed with the results obtained from PCs, as well as new associations between the parameters that were not adequately reported in the previous sections. Strong ($p < 0.01$) and significant correlation ($p < 0.05$) were observed in the groundwater samples. The pH showed a negatively significant correlation with K^+ ($r = -0.54$), Mg^{2+} ($r = -0.57$), Fe^+ ($r = -0.58$), HCO_3^- ($r = -0.44$) respectively with a 95% confidence level. These correlation results indicated mixed source either geogenic or anthropogenic origin, which described in PC1 of the previous part. The K^+, Mg^{2+}, HCO_3^- were the main constituents of groundwater as a result of lithologic influence with leaching of secondary salts by rain water. The EC had a positive strong correlations with Na^+ ($r = 0.58$), Cl^- ($r = 0.69$), HCO_3^- ($r = 0.76$). It is found that the salinity load in groundwater are controlled first by Na^+, and then by Cl^- and HCO_3^-. The existence of strong positive correlation of Na^+ and Cl^- ($r = 0.80$) and weak positive correlation between Ca^{2+} and Mg^{2+} ($r = 0.29$) indicate these parameters have mix sources of the origin. The acidic nature of the groundwater was due to the leaching of altered rocks by acidic rainwater (Edet and Offiong, 2002). These results are similar to PC2. It may be attributed to geogenic sources from the basement rocks or anthropogenic sources of salinity dominance in the study area. A passively significant correlated was existed between Ca with Mn ($r = 0.57$) and HCO_3^- ($r = 0.63$), indicating geogenic origin of groundwater contamination, which was observed in PC3 of the previous section.

Ni exhibited a positive significant correlation with Zn ($r = 0.58$), matching with PC4 in anthropogenic sources, which described in the previous section. As showed a positive significant correlation with F^- ($r = 0.59$), indicating a similar of PC5. Although, major industries were not existence in the study area, small scale ceramics industry and natural activities involved in groundwater chemical alteration. A very low positive correlation was observed between Pb and SO_4^{2-} ($r = 0.04$), indicating a similar of PC6. Agricultural fertilizer and stagnant water may be attributed the main sources of this groundwater hydrochemical evolution in the study area. It can be said that common rock water interaction and anthropogenic activities are responsible for ionic alteration of groundwater in the study area.

3.6. Geostatistical modeling

We investigated the spatial arrangements of data using geostatistical modeling over the study area. We computed the semivariograms models after normalizing the data using ArcGIS (version 10.2). Ordinary kriging was applied for this study. The nugget, the sill, and the range values of the best fitted semivariogram models for quality parameters are presented in Table 8. The predicted and observed values are compared for ordinary kriging interpolation method using the correctness measures to test the robustness of the predicted models (Table 8). The choice of the best semivariogram model is based on the ME, MSE, RMSE, RMSSE, and ASE criterion. A model is considered robust and accurate when the ME and MSE is close to zero, RMSE and ASE are smaller and RMSSE is close to 1 (Adhikary et al., 2010). Cross validation results suggested that all indices values provided more accurate spatial distribution for the study area.

The nugget/sill ratio indicates the spatial dependence of groundwater quality parameters. Three classifications are used to explain the models: the ratio less than 25% indicates strong spatial dependence; the ratio is in between 25 and 75% indicates moderate spatial dependence, and the ratio is more than 75% represents weak spatial dependence (Shi et al., 2007). Fig. 7 is shown that the experimental semivariogram (binned sign) around the omnidirectional semivariogram model (blue line) and average of semivariagram models (plus sign). The exponential semivariagram model identified to be the best fitted models for C_d, HEI, GWQI values, while the circular semivariagrm model fitted best for HPI values (Table 8). These results are consistent with the finding of Munna et al. (2015), who carried out spatial distribution analysis of groundwater quality parameters for Sylhet City Corporation area, Bangladesh. The major ranges varied from 3.33 km to 6.21 km, where the greatest range was measured for C_d, HEI and GWQI (6.21 km), and the smallest one for HPI (3.33 km). The variation of ranges may be related to topographic and geometric factors of groundwater, while large distance and variation of groundwater quality parameters could be affected by small scale

Table 7
Pearson's correlation matrix among physicochemical parameters and metal in the analyzed samples.

	pH	EC	Na^+	K^+	Ca^{2+}	Mg^{2+}	As	Fe	Mn	Ni	Pb	Zn	Cl^-	F^-	HCO_3^-	SO_4^{2-}
pH	1															
EC	−0.218	1														
Na^+	−.298*	.581**	1													
K^+	−.544**	.441**	.400**	1												
Ca^{2+}	0.061	.507**	−0.214	0.05	1											
Mg^{2+}	−.576**	.622**	.312*	.648**	.294*	1										
As	0.234	−0.053	−0.074	−0.164	0.022	−0.115	1									
Fe	−.584**	0.043	.285*	0.184	−.361**	.335**	0.025	1								
Mn	0.087	0.153	−.308*	−0.04	.575**	−0.044	0.049	−.319*	1							
Ni	0.001	.298*	−0.075	0.033	.575**	0.2	−0.066	−0.194	0.237	1						
Pb	−0.026	0.086	0.245	−0.044	0.003	0.104	−0.026	−0.059	−0.01	0.024	1					
Zn	0.232	0.05	−0.109	−0.146	0.161	−0.045	−0.064	−0.159	−0.018	.589**	0.018	1				
Cl^-	−0.102	.693**	.807**	.300*	0.03	.385**	−0.143	0.046	−0.191	0.007	0.194	0.026	1			
F^-	−0.025	−0.009	.339**	−0.056	−.383**	0.027	.590**	.296*	−.388**	−0.11	0.057	−0.002	0.163	1		
HCO_3^-	−.443**	.766**	.423**	.512**	.633**	.725**	−0.045	0.207	0.223	.444**	0.118	−0.002	.337**	−0.069	1	
SO_4^{2-}	−0.032	−0.196	−0.212	−0.044	0.018	−0.04	−0.198	−0.11	0.115	−0.038	0.042	−0.188	−0.155	−0.165	−0.139	1

* Correlation is significant at the 0.05 level (2-tailed).
** Correlation is significant at the 0.01 level (2-tailed).

Table 8
The most suitable characteristic of indices of best fitted semivariogram models for groundwater parameters and their changes.

Indices	Fitted model type	Nugget	Major range	Sill	Nugget/Sill	Lag size	ME	RMSE	MSE	RMSSE	ASE
HPI	Circular	0	3331.76	5924.2	0	416.47	−0.87	54.88	0	0.86	67.39
C_d	Exponential	34.08	6215.78	39.4	0.87	776.97	0.16	5.73	0.02	0.9	6.42
HEI	Exponential	33.43	6215.78	42.63	0.78	776.97	0.17	5.91	0.02	0.9	6.6
GWQI	Exponential	2259.54	6215.78	3476.7	0.65	776.97	1.36	52.75	0.02	0.91	58.24

ME = mean error, RMSE = root mean square error, MSE = mean standardized error, RMSSE = root mean square standardized error, ASE = average standard error.

factors such as precipitation, runoff, and fertilizer application. The ordinary kriging results showed that C_d and HEI values have a weak spatial dependence (Fig. 6), while GWQI represent a moderate spatial dependency (Fig. 7). Only HPI values exhibits a strong spatial dependence (Fig. 7). Most of the groundwater quality parameters were belonging to moderate to strong spatial dependence indicated that less nugget effect in the semivariogram shapes except HPI values.

3.7. Spatial distribution map

The spatial map of GWQI values indicated that high values were observed in northern side of the study area, while poor values were found in the Faridpur Sadar Upazila, Bangladesh (Fig. 8). Poor quality water in Faridpur reason could be happened due to leaching of ions, over exploitation of groundwater, direct discharge of effluents, and agricultural impact (Sahu and Sikdar, 2008; Islam et al., 2015). Islam et al. (2015) reported poor water quality in some sample locations of northern Bangladesh, where slight detrital health effect occurs by using dirking water.

The spatial map of HPI scores demonstrated a complex distribution pattern (Fig. 8). The discrepancies were found in the central part of the study area. Higher values of HPI may be contributed to unsystematic and uncontrolled groundwater withdrawals for domestic purposes. The C_d and HEI exhibit more or less similar distribution patterns with an increasing trend in the northern to southern direction, which suggested the existence of similar point sources (Fig. 8). However, it has been suggested that anthropogenic sources are likely to be attributed the high scores of the HEI and C_d in the study area.

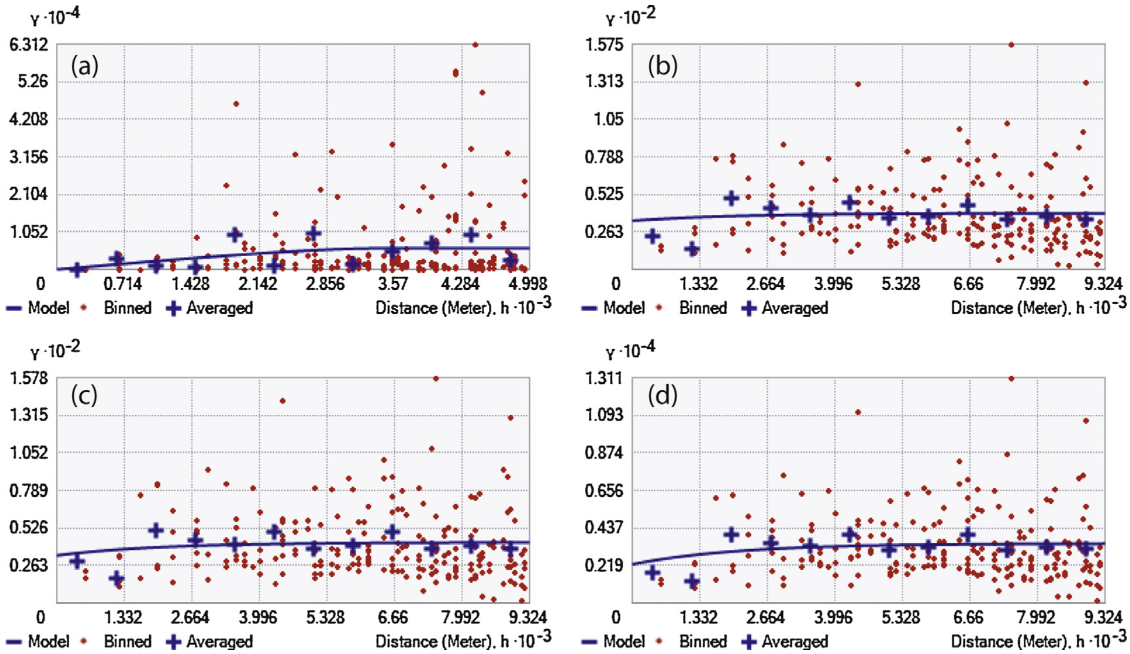

Fig. 7. Best fitted semivariogram model of groundwater quality evaluation indices values in the study area; (a) HPI, (b) C_d, (c) HEI, (d) GQWI.

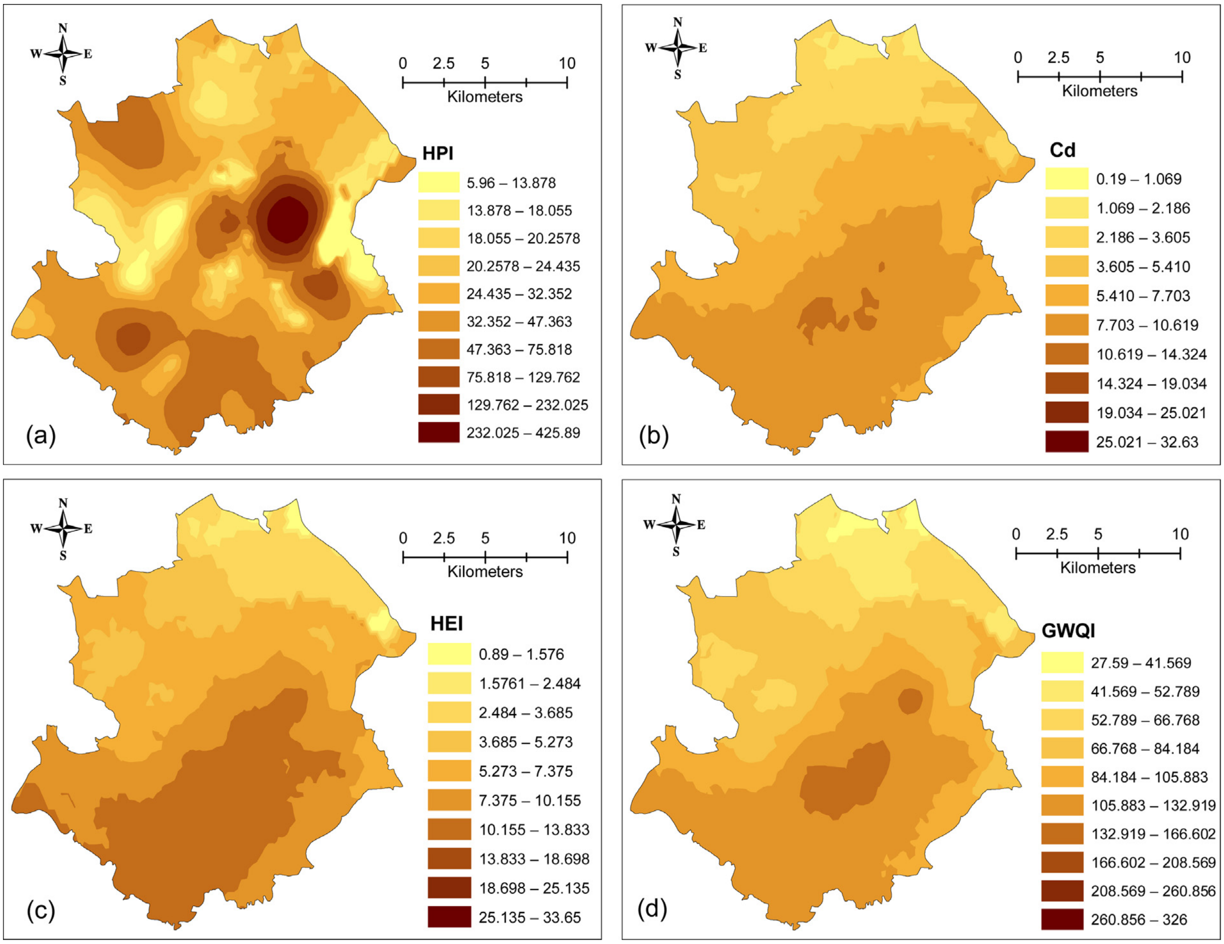

Fig. 8. Maps showing the spatial distribution of four indices scores obtained by quality evaluation indices of the groundwater samples: (a) HPI, (b) C_d; (c) HEI; (d) GWQI.

4. Conclusion

This paper presents integrated approaches for characterizing hydrochemistry and suitability of groundwater quality in the Faridpur district of central Bangladesh. Based on GWQI; about 47% of the samples (28 location) belong to excellent to good water quality type, whereas 52% (29 location) exhibited very poor to poor water quality for drinking purposes in the study area. According to pollution evaluation indices classification; HPI, C_d and HEI showed that 66% (40 locations), 71% (43 locations) and 65% (39 locations) samples reveals low level of pollution in the study area. Although, three schemes of pollution indices were very similar to each other. However, the C_d provided better assessment results. However, spatial analysis of GWQI depicted that high score values for drinking purposes were observed in northern side of the study area, while poor score values exhibited in the middle part of Faridpur region, Bangladesh. The principal component analysis (PCA) demonstrates that anthropogenic (surface runoff, agriculture fertilizers) and natural/geogenic sources (rock–water interaction) are responsible for variation of physicochemical parameters in groundwater aquifer. Besides, the results of cluster analysis (CA) and correlation matrix (CM) support the findings of PCA analysis. The results demonstrate that geogenic processes and anthropogenic sources are controlling factor for affecting groundwater quality in central Bangladesh. The exponential semivariogram model was validated as the best fitted models for C_d, HEI, GWQI values, while circular semivariogram model was fitted best models for HPI value. It is hoped that this study gives adequate background information on physiochemical parameters, water evaluation indices, possible source of pollution, controlling factors of groundwater quality and its spatial distribution in central Bangladesh. This paper is expected to help water resource planners taking adaptive measures for groundwater quality monitoring in central Bangladesh.

Conflict of interest

None.

Acknowledgements

The authors would like to acknowledge Chemistry Division, Atomic Energy Center, Dhaka and Centre for Water Resource Studies, Dalhousie University, Canada for providing some technical support during conducting this study and preparing the manuscript. The authors are grateful to Mohammad Amir Hossain Bhuiyan, Associate Professor, Department of Environmental Sciences, Jahangirnagar University, Bangladesh for his continuous encouragement and kind support for this study.

References

Adhikary, P.P., Chandrasekharan, H., Chakraborty, D., Kamble, K., 2010. Assessment of groundwater pollution in West Delhi, India using geostatistical approach. Environ. Monit. Assess. 167 (1–4), 599–615.

APHA-AWWA-WEF, 2005. Standard Methods for the Examination of Water and Wastewater, 20th ed. APHA, AWWA and WEF, Washington, DC, USA.

Backman, B., Bodis, D., Lahermo, P., Rapant, S., Tarvainen, T., 1997. Application of a groundwater contamination index in Finland and Slovakia. Environ. Geol. 36, 55–64.

Bahar, M.M., Reza, M.S., 2010. Hydrochemical characteristics and quality assessment of shallow groundwater in a coastal area of Southwest Bangladesh. Environ. Earth Sci. (2010) 61, 1065–1073.

Bangladesh Standard (DoE), 1997. The environment conservation rules 1997. Government of the People's Republic of Bangladesh, Dhaka.

Beaulieu, B.T., Savage, K.S., 2005. Arsenate adsorption structures on aluminum oxide and phyllosilicate mineral surfaces in smelter-impacted soils. Environ. Sci. Technol. 39, 3571–3579.

BGS, DPHE, 2001. Arsenic contamination of groundwater in Bangladesh. In: BGS/Technical Report, WC/0019.

Bhuiyan, M.A.H., Suruvi, N.H., Dampare, S.B., Islam, M.A., Quraishi, S.B., Ganyaglo, S., Suzuki, S., 2011. Investigation of the possible sources of heavy metal contamination in lagoon and canal water in the tannery industrial area in Dhaka, Bangladesh. Environ. Monit. Assess. 175, 633–649.

Bhuiyan, M.A.H., Dampare, S.B., Islam, M.A., Suzuki, S., 2015. Source apportionment and pollution evaluation of heavy metals in water and sediments of Buriganga River, Bangladesh, using multivariate analysis and pollution evaluation indices. Environ. Monit. Assess. 187, 4075.

Biswas, R.K., Roy, D.K., Islam, A.R.M.T., Rahman, M.M., Ali, M.M., 2014. Assessment of drinking water related to arsenic and salinity hazard in Patuakhali district, Bangladesh. Int. J. Adv. Geosci. 2 (2), 82–85.

Burrough, P.A., McDonnell, R.A., 1998. Principles of Geographical Information Systems. Oxford University Press, Oxford.

Delhomme, J.P., 1978. Kriging in the hydrosciences. Adv. Water Res. 1, 251–266.

Edet, A.E., Offiong, O.E., 2002. Evaluation of water quality pollution indices for heavy metal contamination monitoring. A study case from Akpabuyo-Odukpani area, Lower Cross River Basin (southeastern Nigeria). GeoJournal 5, 295–304.

ESRI, 2009. ArcGIS Desktop Software. ArcGIS Desktop 9.3, Redlands, CA.

Farnham, I.M., Johannesson, K.H., Singh, A.K., Hodge, V.F., Stetzenbach, K.J., 2003. Factor analytical approaches for evaluating groundwater trace element chemistry data. Anal. Chim. Acta 490, 123–138.

Foster, S.S.D., 1995. Groundwater for development – overview of quality constraints. In: Nash, H., McCall, G.J.H. (Eds.), Groundwater Quality. Water Resources Bulletin, vol. 28, no. 3, pp. 525–533.

Goovaerts, P., 1997. Geostatistics for Natural Resource Evaluation. Oxford University Press, New York, USA.

Gorai, A.K., Kumar, S., 2013. Spatial Distribution Analysis of Groundwater Quality Index Using GIS: A Case Study of Ranchi Municipal Corporation (RMC) Area. Geoinfor. Geostat.: Overview 1, 2.

Gou, X., Li, Y., Wang, G., 2007. Heavy metal concentrations and correlations in rain-fed farm soils of Sifangwu Village, Central Gansu Province, China. Land Degrad. Dev. 18 (1), 77–88.

Halim, M.A., Majumder, R.K., Nessa, S.A., Oda, K., Hiroshiro, Y., Jinno, K., 2010. Arsenic in shallow aquifer in the eastern region of Bangladesh: insights from principal component analysis of groundwater compositions. Environ. Monit. Assess. 61, 453–472.

Han, Y.M., Du, P.X., Cao, J.J., Posmentier, E.S., 2006. Multivariate analysis of heavy metal contamination in urban dusts of Xi'an, Central China. Sci. Total Environ. 355, 176–186.

Holland, H.D., 1978. The Chemistry of the Atmosphere and Oceans. John Wiley and Sons, New York, pp. 351.

Indian Standard, 2012. Bureau of Indian Standards Drinking Water Specifications, BIS 10500:2012, New Delhi, India.

Isaaks, E.H., Srivastava, R.M., 1989. An Introduction to Applied Geostatistics. Oxford University Press, New York, pp. 561.

Islam, A.R.M.T., Rakib, M.A., Islam, M.S., Jahan, K., Patwary, M.A., 2015. Assessment of health hazard of metal concentration in groundwater of Bangladesh. Am. Chem. Sci. J. 5 (1), 41–49, http://dx.doi.org/10.9734/ACSj/2015/13175.

Liu, C.W., Lin, K.H., Kuo, Y.M., 2003. Application of factor analysis in the assessment of groundwater quality in a blackfoot disease area in Taiwan. Sci. Total Environ. 313, 77–89.

Marko, K., Al-Amri, N.S., Elfeki, A.M.M., 2014. Geostatistical analysis using GIS for mapping groundwater quality: case study in the recharge area of Wadi Usfan, western Saudi Arabia. Arab. J. Geosci. 7, 5239–5252, http://dx.doi.org/10.1007/s12517-013-1156-2.

Masoud, A.A., 2014. Groundwater quality assessment of the shallow aquifers west of the Nile Delta (Egypt) using multivariate statistical and geostatistical techniques. J. African Earth Sci. 95, 123–137.

McCutcheon, S.C., Martin, J.L., Barnwell, T.O., 1993. Water quality. In: Maidment, D.R. (Ed.), Handbook of Hydrology. McGraw-Hill, Inc., New York, pp. 11.1–11.73.

McKenna Jr., J.E., 2003. An enhanced cluster analysis program with bootstrap significance testing for ecological community analysis. Environ. Model. Softw. 18 (3), 205–220.

Meharg, A.A., Rahman, M.M., 2003. Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption. Environ. Sci. Technol. 37, 229–234.

Mendiguchıía, C., Moreno, C., Galindo, R.M.D., Garcıí a-Vargas, M., 2004. Using chemometric tools to assess anthropogenic effects in river water. A case study: Guadalquivir River (Spain). Anal. Chim. Acta 515, 143–149.

Mohan, S.V., Nithila, P., Reddy, S.J., 1996. Estimation of heavy metals in drinking water and development of heavy metal pollution index. J. Environ. Sci. Health Part A 31, 283–289.

Molla, M.A., Saha, N., Salam, S.A., Rakib-uz-Zaman, M., 2015. Surface and groundwater quality assessment based on multivariate statistical techniques in the vicinity of Mohanpur, Bangladesh. Int. J. Environ. Health Eng. 4, 18, http://dx.doi.org/10.4103/2277-9183.157717.

Munna, G.M., Kibriya, N.A., Nury, A.H., Islam, S., Rahman, H., 2015. Spatial distribution analysis and mapping of groundwater quality parameters for the Sylhet City Corporation (SCC) area using GIS. Hydrology 3 (1), 1–10.

Nabizadeh, R., Maryam, V.A., Mahmood, A., Kazem, N., Amir, H.M., Samira, Y., 2013. Development of innovative computer software to facilitate the setup and computation of water quality index. J. Environ. Health Sci. Eng. 11, 1, http://www.ijehse.com/content/11/1/1.

Omo-Irabor, O.O., Olobaniyi, S.B., Oduyemi, K., Akunna, J., 2008. Surface and groundwater water quality assessment using multivariate analytical methods: a case study of the Western Niger Delta, Nigeria. Phys. Chem. Earth 33, 666–673.

Prasad, B., Bose, J.M., 2001. Evaluation of the heavy metal pollution index for surface and spring water near a limestone mining area of the lower Himalayas. Environ. Geol. 41, 183–188.

Prasad, B., Jaiprakas, K.C., 1999. Evaluation of heavy metals in ground water near mining area and development of heavy metal pollution index. J. Environ. Sci. Health A34, 91–102.

Quddus, K.G., Zaman, M.W., 1996. Irrigation water quality in some selected villages of Meherpur in Bangladesh. J. Agric. Sci. 23 (1), 51–57.

Rahman, M.A.T.M.T., Rahman, S.H., Majumder, R.K., 2012a. Groundwater quality for irrigation of deep aquifer in southwestern zone of Bangladesh. Songklanakarin J. Sci. Technol. 34 (3), 345–352.

Rahman, M.A.T.M.T., Saadat, A.H.M., Islam, M.S., Al-Mansur, M.A., Ahmed, S., 2014a. Groundwater characterization and selection of suitable water type for irrigation in the western region of Bangladesh. Appl. Water Sci., http://dx.doi.org/10.1007/s13201-014-0239-x.

Rahman, M.S., Gagnon, G.A., 2014. Bench-scale evaluation of drinking water treatment parameters on iron particles and water quality. Water Res. 48, 137–147.

Rahman, M.S., Molla, A.H., Saha, N., Rahman, A., 2012b. Study on heavy metals levels and its risk assessment in some edible fishes from Bangshi River, Savar, Dhaka, Bangladesh. Food Chem. 134, 1847–1854.

Rahman, M.S., Saha, N., Molla, A.H., 2014b. Potential ecological risk assessment of heavy metal contamination in sediment and water body around Dhaka export processing zone, Bangladesh. Environ. Earth Sci. 71 (5), 2293–2308.

Rahman, M.S., Whalen, M., Gagnon, G.A., 2013. Adsorption of dissolved organic matter (DOM) onto the synthetic iron pipe corrosion scales (goethite and magnetite): effect of pH. Chem. Eng. J. 234, 149–157.

Raihan, F., Alam, J.B., 2008. Assessment of groundwater quality in Sunamganj Bangladesh. Iranian J. Environ. Health Sci. Eng. 6 (3), 155–166.

Reza, A.H.M.S., Jean, J.S., Lee, M.K., Liu, C.C., Bundschuh, J., Yang, H.J., Lee, J.F., Lee, Y.C., 2010. Implications of organic matter on arsenic mobilization into groundwater: evidence from northwestern (Chapai-Nawabganj), central (Manikganj) and southwestern (Chadpur) Bangladesh. Water Res. 44, 5556–5574.

Sahu, P., Sikdar, P.K., 2008. Hydrochemical framework of the aquifer in and around East Kolkata Wetlands, West Bengal, India. Environ. Geol. 55, 823–835.

Shahid, S.U., Iqbal, J., Hasnain, J., 2014. Groundwater quality assessment and its correlation with gastroenteritis using GIS: a case study of Rawal Town, Rawalpindi, Pakistan. Environ. Monit. Assess. (2014) 186, 7525–7537, http://dx.doi.org/10.1007/s10661-014-3945-9.

Shahidullah, S.M., Hakim, M.A., Alam, M.S., Shansuddoha, A.T.M., 2000. Assessment of groundwater quality in a selected area of Bangladesh. Pak. J. Biol. Sci. 3 (2), 246–249.

Shi, J., Wang, H., Xu, J., Wu, J., Liu, X., Zhu, H., Yu, C., 2007. Spatial distribution of heavy metals in soils: a case study of Changxing, China. Environ. Geol. 52 (1), 1–10.

Singh, P.K., Tiwari, A.K., Panigarhy, B.P., Mahato, M.K., 2013. Water quality indices used for water resources vulnerability assessment using GIS technique: a review. Int. J. Earth Sci. Eng. 6 (6–1), 1594–1600.

Tapoglou, E., Karatzas, G.P., Trichakis, J.C., Varouchakis, E.A., 2014. A spatio-temporal hybrid neural network-Kriging model for groundwater level simulation. J. Hydrol. 519, 3193–3203.

Tiwari, A.K., Singh, P.K., Mahato, M.K., 2014. GIS-based evaluation of water quality index of groundwater resources in west Bokaro Coalfield, India. Curr. World Environ. 9 (3), 73–79.

Varouchakis, E.A., Hristopulos, D.T., 2013. Improvement of groundwater level prediction in sparsely gauged basins using physical laws and local geographic features as auxiliary variables. Adv. Water Resour. 52, 34–49.

Vasanthavigar, M., Srinivasamoorthy, K., Vijayaragavan, K., Ganthi, R.R., Chidambaram, S., Anandhan, P., Manivannan, R., Vasudevan, S., 2010. Application of water quality index for groundwater quality assessment: Thirumanimuttar sub-basin, Tamilnadu, India. Environ. Monit. Assess. 171 (1–4), 595–609, http://dx.doi.org/10.1007/s10661-009-1302-1.

Webster, R., Oliver, M., 2001. Geostatistics for Environmental Scientists. John Wiley & Sons, Ltd, Chichester, pp. 271.

WHO, 2004. Guidelines for Drinking-Water Quality, 3rd ed. WHO, Geneva, Switzerland.

WHO, 2011. Guidelines for Drinking-Water Quality, 4th ed. World Health (WHO) Organization.

Zaman, M.W., Nizam, M.A., Rahman, M.M., 2001. Arsenic and trace element toxicity in groundwater for agricultural, drinking and industrial usage. Bangladesh J. Agric. Res. 26 (3), 167–177.