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ASSESSMENT OF CONTAMINATION POTENTIAL OF GROUNDWATER ADJACENT TO THE WASTE DISPOSAL SITE AT KHULNA: CONTAMINATION INDICES, MULTIVARIATE STATISTICS AND GEOSTATISTICAL APPROACH

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ABSTRACT

The main focus of this study was to evaluate the contamination potential of groundwater using various contamination indices available in the literature. To these attempts, fifteen water samples were collected from selected production well located nearby waste disposal site at Rajbandh, Khulna, Bangladesh. In the laboratory, the concentrations of heavy metals of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, Na, K, Ca, As, Mg in water were measuredthrough standard test methods. The groundwater contamination indices such as groundwater quality index (GWQI), degree of contamination (C_d), heavy metal pollution index (HPI) and heavy metal evaluation index (HEI) were used to quantify the level of contamination. In this study, for evaluating the level of contamination, the standard limit and ideal values of groundwater were obtained from BIS (10500:2012) and BDS (1240:2001), respectively. Numerous researchers published various limits for GWQI, C_d, HPI and HEI for categorized the level of contaminations such as excellent water, good water, poor water and very poor water. In this study, result of GWQI reveals that 26.67% of total water samples belong to very poor, while, 73.33% belong to poor water. The correlation between these four indices indicated that GWQI and C_d provided better results with consistent value.Result of Pearson's correlataion depicts that most of the heavy metals were barely correlated.In addition, results of PCA indicated that As in groundwater was contaminated from anthropogenic activities, while, Na, Ca and Mg were from natural sources. The heavy metals and various indices were distributed spatially. Finally, it is expected that outcomes of this study will provide a guideline for decision makers taking proper measures for groundwater quality management all over the world.

Keywords:*Disposal site, groundwater, heavy metal, contamination indices, PCA.*

1.INTRODUCTION

Waste disposal site or Landfilling is widely used inmunicipal solid waste (MSW) management practices in developing countries (Mangimbulude, Breukelen, Krave, Straalen, & Röling, 2009).landfills have been the most common method of organized MSW disposal and remain so in many places around the world.MSW landfill is a method of disposing of refusal on land by utilizing the principles of engineering. Leachate, a toxic liquid generated in municipal landfill contains large amounts of organic and inorganic contaminants by means of physical, chemical and microbiological changes. The leachate will continuously migrate through the soil strata and eventually the groundwater systemthat have been contaminated with heavy metals such as lead, copper, zinc, iron, manganese, chromium, cadmium and these heavy metals in MSW lead to serious problems because they cannot be bio-degraded. Khulna, the third largest city of Bangladesh is located at the south-western part of the country. It has an increasing growth rate of 5% having total population of 2.3 million in Khulna zilla(BBS, 2011).In addition, water is the basic and one of the vital physical components to run the livelihood. But safe and adequate sustainable drinking water is all that the mankind need. With an increasing rate of this population, demand of drinking water also increases. River, pond, rain, and groundwater are the main source of water. But, in coastal region like Khulna, salinity of water induced many problem. Rivers in Khulna region are established as polluted(Sabbir et al., 2010; Ahmed et al., 2015). It also includes heavy metal contamination (Kibria, Hossain, Mallick, Lau, & Wu, 2016). Groundwater is a valuable renewable resource and usually microbiologically safe and chemically stable in the absence of direct contamination(WHO, 2011). The quality of groundwater primarily depends on geological formation of a particular region as well as anthropogenic or human activities. In this study, fifteen water samples were collected from production well located nearby waste disposal site at Rajbandh, Khulna, Bangladesh. In te laboratory, the concentrations of relevant heavy metals of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, Na, K, Ca, As, Mg in water samples were measured through standards test methods. The groundwater contamination indices such as groundwater quality index (GWQI), degree of contamination (C_d), heavy metal pollution index (HPI) and heavy metal evaluation index (HEI) were used to quantify the level of contamination.

2.MATERIALS AND METHODS

2.1.Study Area

Khulna is the south-western sea-adjacent third largest city in the geography of Bangladesh on the banks of Rupsha and Bhairabriver. It’s located at 22°49′0″N 89°33′0″E with a total area of 4394.46 km^2. The selected disposal site at Rajbandh(Figure 1)is a place 7 km north from Khulna city center which is currently used for open filling of MSW.The waste disposal site i.e. causes contamination of surrounding surface water bodies, groundwater sources and underlying soil layer. For drinking and irrigation purpose groundwater is the main source in Khulna division and that’s why toxicity control in such area is a mandatory work to do.

Figure 1: Water sampling location from production wells nearby

2.2. Collection of groundwater

Before collecting water samples the bottle was washed by distilled water several times. Then the bottles were air or sun dried. Then 2-3 mL a solution was used as preservative. The preservative was prepared by mixing concentrated nitric acid and distilled water at a ratio of 1:1. Then the bottle was kept for 24 hours at room temperature. After that the bottles were prepared for collecting water sample. In this study, fifteen groundwater samples were collected from selected production wells or tube wells located adjacent to the waste disposal site at Rajbandh of Khulna, Bangladesh. Moreover, fifteen surface water samples were collected from pond located at the mentioned locations. All the sampling points were gathered with the help of GPS shown in Figure 1. These study periods covered both the dry and rainy seasons.

2.3. Laboratory Investigations

Both the water samples were collected from the site and then brought to DPHE, Khulna, Bangladesh. The concentrations of heavy metals of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, Na, K, Ca, As in water were measured through atomic absorption spectrophotometer (AAS).

2.4.Drinking water evaluation indices

To evaluate the level of contamination of groundwater, various indices such as groundwater quality index (GWQI), degree of contamination (C_d), heavy metal pollution index (HPI) and heavy metal evaluation index (HEI) proposed by different researchers were used and hence discussed in the following articles.

2.4.1. Ground water quality index:

GWQIhelps to determine the acceptability of drinking water.Groundwater quality of the collected water samples from different production well nearby waste disposal site was investigatedusing the following Equation 1.

$$GWQI = \sum SI_i = \sum(W_i \times q_i) = \sum[\left(\frac{w_i}{\sum_{n}^{i=1} w_i}\right) \times \left(\frac{C_i}{S_i} \times 100\right)] \quad (1)$$

Where C_i is the concentration of each heavy metal, S_i is the limit values, $w_i$is the assigned weightage, q_i is water quality rating, W_i is the relative weight, $SI_i$is the sub-index of *i*thheavy metal. In this study, the weight factors and limit values proposed by (Nabizadeh et al., 2013)and (Vasanthavigar et al., 2010) were considered for evaluating GWQI provided in Table 1

Table 1: Parameters, weight factors and limit values considered for GWQI

Parameter	Units	Weight factor (w_i)	Relative Weight (W_i)	Limit value(s_i), BIS
Fe	mg/l	4	0.10	0.300
Mn	mg/l	4	0.10	0.3
Cr(+6)	mg/l	4	0.10	0.05
Cu	mg/l	2	0.05	1.5
Pb	mg/l	4	0.10	0.01
Zn	mg/l	3	0.07	5
Ni	mg/l	3	0.07	0.1
Cd	mg/l	4	0.10	0.005
Na	mg/l	4	0.10	200
K	mg/l	2	0.05	12
Ca	mg/l	2	0.05	200
As	mg/l	4	0.1	0.01
Mg	mg/l	2	0.05	100
	$\sum w_i =$	42	$\sum w_i = 1.00$	

2.4.2.Heavy metal pollution index(HPI)

Heavy metal pollution index (HPI), is a rating method that qualify water quality with respect to heavy metals by assigning unit weightage (W_i). The unit weightage (W_i) is defined inversely proportional to the standard value (S_i) (Mohan et al.,1996).In this study, HPI was computed using the following Equation (2).

$$HPI=\frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \quad (2)$$

Where, $Q_i$is the sub-index of the *i*th parameter and $W_i$is unit weight of the *i*th parameter and *n* is the number of parameters. The sub-index $Q_i$is computed byEquation (3)

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

Where, M_i, I_i, and $S_i$denote 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. Moreover, the unit weightage $W_i$was computedusing following Equation (4).

$$W_i = \frac{K}{S_i} \quad (4)$$

Where K is proportional constant.
In this study, the concentration limits i.e., standard value (S_i) and ideal value (I_i) for each heavy metal in water were taken from BIS (2012) andBDS (1240:2001), respectively.

2.4.3.Heavy metal evaluation index(HEI)

Heavy metal evaluation index (HEI) is a method of evaluating water quality parameter contaminated with heavy metal and interpret a thorough investigation of the level of contamination in groundwater(Prasad & Jaiprakas, 1999).In this study, HEI was calculated using the following Equation (5).

$$HEI=\sum_{i=1}^{n} \frac{H_c}{H_{mac}} \quad (5)$$

Where, H_c and H_{mac} are monitored and maximum admissible value, respectively.

2.4.4.The degree of contamination (C_d)

The degree of contamination of groundwater was computed fromthe following Equation (6)(Backman, Bodiš, Lahermo, Rapant, & Tarvainen, 1998),

$$C_d = \sum_{i=1}^{n} C_{fi} (6)$$

Where, $Cf_i = (\frac{C_{ai}}{C_{ni}}) - 1$ (6)

Where,$Cf_i$is the contamination factor, $C_{ai}$and $C_{ni}$are the analytical value and upper permissibleconcentration for the *i*th component respectively, and *n* is indicated for the normative value. Here, $C_{ni}$was taken as maximum permissible concentration

2.5.Multivariate Statistical Analysis

In this study, Pearson's correlation, principal component analysis (PCA) and cluster analysis (CA) were performed using XLSTAT. Pearson correlation was carried out in this study to demonstrate the correlation coefficient matrix of the water quality data in which coefficients of correlation data was calculated using the following Equation (7).

$$r = \frac{\sum(xy)-(\sum x)(\sum y)}{\sqrt{[n(\sum x^2)-(\sum x)^2][n(\sum y^2)-(\sum y)^2]}} \quad (7)$$

2.6.Geo-statistical Modeling

For Geostatic Modeling, Ordinary Kriging(OK) was performed using ArcGIS as an optimal interpolation based on regression against observed values of surrounding data points, weighted according to spatial covariance values(Bohling, 2005).The spatial variation can be illustrated by thefollowing Equation (8) proposed byDelhomme (1978).

$$\hat{z}(X_o) = \sum_{i=1}^{n} \lambda_i Z(x_i) \quad (8)$$

Where $\hat{z}$ is the Estimated value of an attribute at the point of interest x_o, z is the observed value at the sample point x_i, n is the number of sampled points, λ_i is the weight assigned to the sampled point.

3.RESULTS AND DISCUSSION

The results of groundwater quality, various indices, the correlations and the spatial distribution of heavy metals are presented and hence discussed in the following articles.

3.1.General Characteristics of Groundwater Quality

The descriptive statistics such as maximum, minimum mean, standard deviation (SD) and variance of heavy metal in groundwater of production well nearby disposal site are provided in Table 2. The values of SD indicates how much the parameters deviates from the mean value. In this study,the maximum SD was found for Ca (33.88218) denoted that the concentration of Ca was spread widely from the mean value of it. The heavy metal of Ni have the lowest SD (0.010898) indicated that it is closely related to the mean value. The variance is the squared numerical value of SD of the randomly selected parameter. In this study maximum and minimum variance follow standard deviation of parameters respectively.

Table 2: Descriptive statistics of physiochemical parameters of heavy metal in study area

Parameter	Maximum	Minimum	Mean	Std. Deviation	Variance	BIS (2012)	BDS (2001)
Fe	2.3	0.01344	0.426193	0.539286	0.290830	0.3	1
Mn	0.352	0.016	0.219326	0.299833	0.089900	0.3	0.1
Cr (+6)	0.135648	0.007	0.040888	0.024517	0.000601	0.05	0.01
Cu	0.95	0.34	0.554938	0.162604	0.026440	1.5	0.05
Pb	0.05755	0.02	0.02598	0.012682	0.000160	0.01	0.05
Zn	1.8	0.029	0.576960	0.542613	0.294429	5	3
Ni	0.089	0.033	0.052305	0.010898	0.000118	0.1	0.02
Cd	0.07	0	0.142028	0.177738	0.031590	0.005	0.003
Na	120	8	31.82222	29.55271	873.3627	200	150
K	30.83	11	15.52592	4.207834	17.70587	12	10
Ca	258	13	40.36790	33.88218	1148.002	200	75
As	0.045	0.002	0.013889	0.01270	0.000161	0.01	0.05
Mg	116	43	56.87511	7.976963	63.63193	100	30

3.2.Evaluation of Drinking Water Quality

Ground water quality index is used for rating the suitability of ground water for drinking purpose (Abbasi & Abbasi, 2012). (BIS, 2012) and BDS (2001) have given the maximum permissible concentration of the selected parameter.Vasanthavigar(2010) showed a procedure for measuring the water quality index in 15 sampling points having GWQI values ranges from 114.338 to 226.535 with a mean of 182.308 (Table 3). GWQI Result reveals that 26.67% of total water samples (GW2, GW3, GW4, GW5) belong to very poor water as well as remaining 73.33% of total water samples belong to poor water (GW1, GW6, GW7-15). C_d refers to the minimum, maximum and mean value of pollution level i.e. 0.140, 12.196 and 7.544 respectively (Table 4).HEI ranges from 2.77 to 255.13. In this study based on HEI 73.33% of samples were less polluted water, 6.67% of samples contain medium polluted water and 20%

of them is high polluted water.From the comparing perspective, the three classification kinds GWQI, HEI, C_d gives a similar result regarding water quality in Khulna region i.e. poor or very poor quality of water where the HPI value announced that the sample water is not suitable for drinking purpose. However among the four indices, GWQI & C_d interprets good result, marks the ground water as poor and very poor water but not unsuitable for drinking at all.

Table 3: Groundwater indices evaluation: Drinking water quality indices.

Sample ID	**GWQI**	**C_d**	**HEI**	**HPI**
GW1	157.33	5.017488	3.21652	502.122
GW2	203.66	9.798888	37.4223	1016.86
GW3	226.53	12.19627	2.98990	1315.00
GW4	220.77	11.53266	2.77199	1341.08
GW5	220.77	11.53	255.133	1341.08
GW6	114.34	0.14	126.039	67.7539
GW7	160.69	5.015611	4.71165	920.573
GW8	190.04	8.791011	4.58955	889.238
GW9	193.13	8.674593	4.53077	913.778
GW10	198.28	9.218133	3.70521	1251.90
GW11	159.48	5.049558	17.7109	838.600
GW12	164.70	5.562125	3.84595	983.282
GW13	122.03	1.336918	5.79016	295.899
GW14	197.45	9.1897201	2.99868	988.249
GW15	205.41	10.10532	3.88761	1141.37
Minimum	114.34	0.14	2.77199	67.7539
Maximum	226.53	12.19627	255.13	1341.08
Mean	182.31	7.54	31.96	920.45

Table 4: Classification of the groundwater quality of the study area based on modified categories of quality indices value.

Index Method	**Category**	**Degree of pollution/ Water Class**	**Number of locations**	**% of Sample**	**Sample ID**
HPI	<45	Low	0	0	
	45-90	Medium	1	6.67	GW6
	>90	High	14	93.33	GW1, GW2, GW3, GW4, GW5, GW7, GW8, GW9, GW10, GW11, GW12, GW13, GW14, GW15
HEI	<10	Low	11	73.33	GW1, GW3, GW4, GW7, GW8, GW9, GW10, GW12, GW13, GW14, GW15
	10-20	Medium	1	6.67	GW11
	>20	High	3	20	GW2, GW5, GW6
Cd	<10	Low	12	80	GW1, GW2, GW6, GW7, GW8, GW9, GW10, GW11 GW12, GW13, GW14, GW15
	10-20	Medium	3	20	GW3, GW4, GW5
	>20	High	0	0	

GWQI	<50	Excellent water	0	0	
	50-100	Good Water	0	0	
	100.1-200	Poor water	11	73.33	GW9, GW10, GW11, GW12, GW13, GW14, GW15
	200.1-300	VeryPoor water	4	26.67	GW2, GW3, GW4, GW5
	>300	Not suitable for drinking water	0	0	

Pollution source and factors affecting ground water quality

Multivariate statistical techniques e.g. correlation analysis, principle component analysis (PCA) and cluster analysis (CA) are commonly used in environmental engineering studies to understand the sources of pollutants better(Mendiguchía, Moreno, Galindo-Riaño, & García-Vargas, 2004). PCA obstains observation vs. factors graph and variables vs. factors graph. In this study F1 & F2 indicates anthropogenic and natural sources of pollution. The graphical representation of observation vs. factors shows that GW5, GW11 are polluted by antropogenicsources where natural sources pollutes the GW6,GW12,GW7 sample points. GW1,GW2,GW8 are polluted by more or less of both sources as their points are significantly far from the factors (Figure 2). From the variables vs. factors graph it can be seen that the heavy metal of As contamination occurs due to anthropogenic pollution and the Na, Ca, Mg pollution is the result of natural pollution. Cu, K, Cr pollution of site are strongly inter-related based on the sources of pollution as they rely closly on the graph (Figure 2).

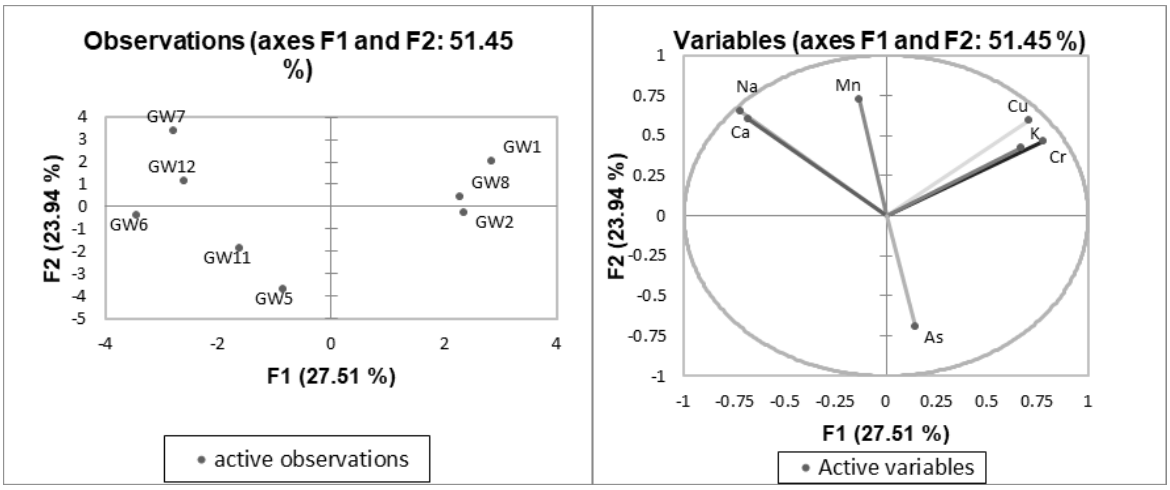

Figure2: PCA Analysis for active observation and active variables

Table 5: Varimax rotated principal component analysis for groundwater samples.

SiteQ mode	F1	F2	` F3	F4	F5
GW1	2.842	2.047	-1.053	-2.019	-0.737
GW2	2.348	-0.264	-0.219	-0.628	0.358
GW3	1.687	-0.639	1.271	1.248	-0.903
GW4	0.213	-2.167	1.061	0.830	0.648
GW5	-0.846	-3.673	1.084	-2.526	-0.260
GW6	-3.432	-0.373	-2.866	-0.001	1.142
GW7	-2.777	3.387	2.826	-0.052	-0.198
GW8	2.279	0.434	-0.096	-0.320	0.213
GW9	1.362	0.515	-0.990	1.406	-1.139
GW10	0.010	-1.249	-0.583	1.200	-0.721
GW11	-1.618	-1.854	-0.645	0.766	-1.228
GW12	-2.606	1.142	0.244	-0.813	-1.409
GW13	-0.332	2.117	-1.636	-0.107	0.660

GW14	0.523	0.924	0.722	1.025	1.052
GW15	0.347	-0.346	0.881	-0.008	2.522

3.3.Spatial similarity and sampling sites grouping

Q-mode Cluster analysis was performed to establish the dendrogram of source-based similarities among disposal site and the factor points. This Dendrogram (Figure 3) denotes the similarities in sources with respect to experimented parameters in a sample cluster. (Bodrud-Doza et al., 2016).

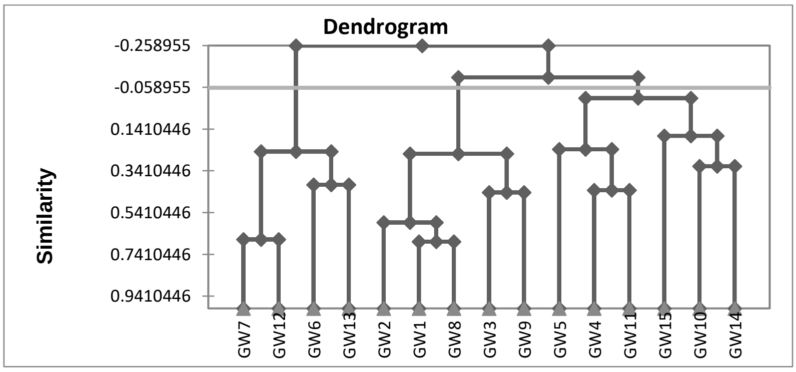

Figure 3: Dendrogram of similarity of parametersand sample locations

The 15 sampling site categorized into three clusters; cluster 1 includes 5 sampling points which areGW1-3, GW8-9. Six sample points fall into cluster 2 category and they are GW4-5, GW10-11, GW14-15. Cluster 3 have 4 sampling points at GW6-7 and GW12-13. Cluster categories express the similarity of pollution sources among the sampling points. Main sources of contamination are agricultural fertilizer, domestic sewage drainage, leaching of parent materials, agricultural runoff and so on. The contamination is grouped of two kinds e.g. natural sources and human activity sources.

3.5.Spatial distribution map

From Arc GIS, various types of spatial distribution was found from different indices which is shown in Figure 4. As for GWQI, the Eastern part of the landfill area shows higher values than the western area which denotes poor water and very poor water respectively. On the other hand, northern part of the landfill shows better quality of water than the southern part. As for C_d, it shows a lot similar distribution to the GWQI. This poor quality of water could be the reason for leaching of ions as well as discharge of wastes or agricultural impacts too (Sahu & Sikdar, 2008). Although the distribution of HPI and HEI shows a complex distribution pattern, the distribution of them shows more or less similar pattern. For both distribution, southern part of the area shows very poor water quality than the other sides. Anthropogenic sources may be responsible for this high number of HEI and HPI.

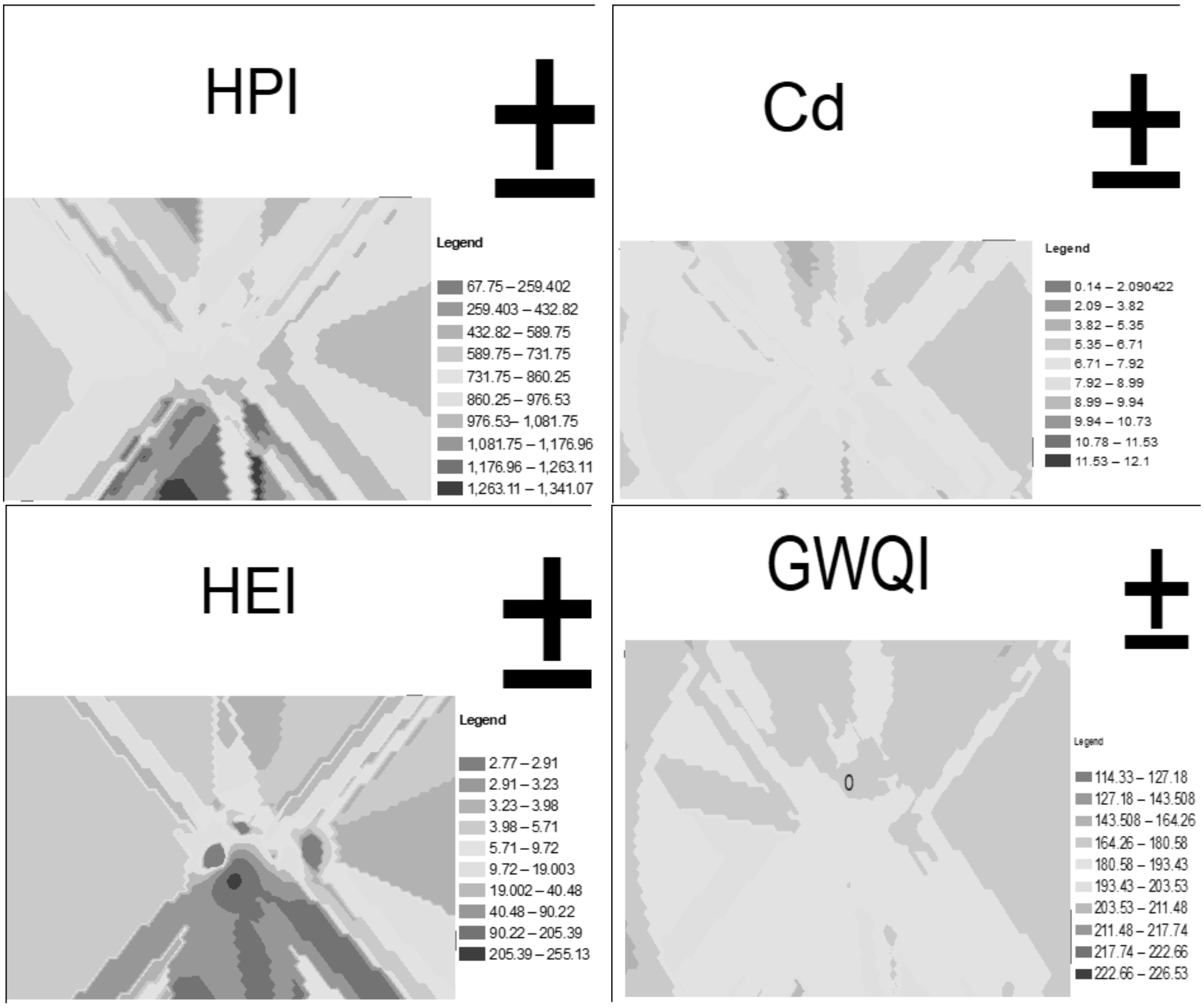

Figure 4: Spatial Distribution map for Four indices near Rajbandh, Khulna.

3.6.Correlation matrix analysis

Pearson's correlation was used to measure the interrelationship and coherence pattern among of groundwater quality parameters. Correlation matrix(Table 6) showed inter-parameter relationships agreed with the results obtained from XLSTAT reported with a 95% confidence level.These correlation results indicated mixed source either anthropogenic origin or geogenic, which was identified by F1 and F2 respectively.Anthropogenic sources dominant due to waste disposal.Whereasnatural activities involved in groundwater chemical alteration.Waste disposal may be attributed the main sources of this groundwater hydro chemical evolution in the study area.The Pearson’s correlation matrix of metal elements in groundwater is provided in Table 6. The most significant correlation was observed for Na and Ca (0.938). However, the concentrations of Fe showed very weak positive correlations with Cr (0.005). This indicated that Fe was from different sources than Cr (Yao, Li, Xie, & Yu, 2012). Based on the results of Pearson’s correlations matrix on waste disposal site, it was observed the high positively correlations between Na and Ca (0.938), Cr and Cu (0.831), Ca and Mg (0.714), Cu and K (0.675), Pb and K (0.632) as well as Mn and Na (0.534) in Table 6.

Table 6: Pearson's correlation matrix of heavy metals in water of wate disposal site

	Fe	Mn	Cr	Cu	Pb	Zn	Ni	Cd	Na	K	Ca	As	Mg
Fe	**1**												
Mn	-0.158	**1**											
Cr	0.005	0.204	**1**										
Cu	0.245	0.317	0.831	**1**									
Pb	0.249	0.086	0.547	0.500	**1**								
Zn	-0.127	0.130	-0.281	-0.009	-0.002	**1**							
Ni	-0.067	-0.414	-0.417	-0.537	-0.052	0.075	**1**						
Cd	-0.067	-0.094	0.023	0.131	0.209	0.056	-0.102	**1**					
Na	-0.306	0.534	-0.326	-0.160	-0.170	0.346	0.055	-0.016	**1**				
K	0.251	-0.036	0.651	0.675	0.632	-0.147	-0.055	0.442	-0.152	**1**			
Ca	-0.207	0.330	-0.268	-0.116	-0.131	0.331	0.064	-0.119	0.938	-0.111	**1**		
As	0.431	-0.581	-0.285	-0.293	0.060	0.108	0.108	-0.131	-0.500	-0.051	-0.390	**1**	
Mg	0.081	0.211	-0.072	0.020	0.164	0.212	0.342	-0.246	0.627	0.269	0.714	-0.154	**1**

4.CONCLUSIONS

In this study, to evaluate the level of contamination of groundwater collected from different selected production well located adjacent to the waste disposal site, various indices of GWQI,Cd,HPI and HEI were used.Results of GWQI reveals that 26.67% of total water samples (GW2, GW3, GW4, GW5) belong to very poor water, while, remaining 73.33% of total water samples belong to poor water (GW1, GW6, GW7-15). In addition, C_d refers to the minimum, maximum and mean value of contamination level i.e. 0.140, 12.196 and 7.544 respectively.HPI ranges from 67.76 to 1341.08 with a mean value of 920.45.In this study based on HEI 73.33% of samples were less polluted water, 6.67% of samples contain medium polluted water and 20% of them is highly polluted. From comparing perspective of these indices, GWQI and C_d indicated that the quality of water of production wells were poor and very poor condition respectively but not unsuitable for drinking purposes. The results of Pearson's correlations indicated that most of the heavy metals had highly positive correlations. Furthermore, results of PCA indicated that the contamination of As was from anthropogenic activities, while, Na, Ca and Mg from natural sources. The spatial distribution of various indices reveals that the contamination of water was found comparatively higher in the production well located near the disposal site as well as decreases in relation to the increasing of water sampling distances.

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