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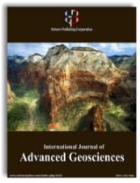

Assessment of heavy metals contamination of the water resources in the vicinity of Barapukuria Coal Mine, Phulbari, Dinajpur, NW Bangladesh

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Abstract

The present research work shows that the concentrations of heavy metals can be expressed as Fe>Mn>Zn>Ni>Cr>Pb>Co>As in the groundwater of Phulbari area, Dinajpur, Bangladesh on the basis of their abundance. The Plio-Pleistocene Dupi Tila aquifer sediment is principally composed of quartz, feldspar and lithic grains and is characterized by low ZTR index. Seventeen groundwater samples have higher iron concentrations than the permissible limit of WHO, 2004. The chemical weathering of pyrite release iron in the natural water of the study area. The manganese content of two groundwater samples is higher than the acceptable limit. The high abundance of potassium in 11.11% water samples is indicative of the influence of agricultural activity in the investigated area. The factor analysis reveals the dominance of four factors that comprise 77.276% of the groundwater sample. The contamination index (Cd) for the heavy metals were performed and 88.89% of the groundwater samples have low contamination index. The calcium of the groundwater is resulted from the dissolution of carbonate rocks. The low arsenic concentrations indicate the fast flowing nature of the groundwater and/or low arsenic content of the aquifer sediments. The low cobalt content of the groundwater reflects the low cobalt concentration of the aquifer sediments. The heavy metal chemistry of the groundwater of the study area reveals that the quality of the water is influenced by the geogenic sources as well as the anthropogenic activities like coal mining and agricultural activities.

Keywords: *Factor analysis; Contamination Index (Cd); Iron; Arsenic; Pyrite and ZTR Index.*

1. Introduction

Groundwater is a natural resource that is necessary for the economic development of a country. Groundwater is used for industrial, domestic and agricultural activities. The study of groundwater quality is a regular routine work for the proper management of the groundwater. The groundwater may be contaminated from various activities and they can be categorized either geogenic or anthropogenic. The present research work deals with the assessment of the groundwater quality of Phulbari Upazila, Dinajpur, Bangladesh where the groundwater can be contaminated principally from the mining and agricultural activities.

The study of the stratigraphic succession of the Barapukuria Basin shows that the coal bearing Gondwana Group overlies on the Precambrian basement complex (Hossain 1999). Hossain et al 2017 summarized that the minerals like illite-smectite, kaolinite, chlorite, sericite, laumontite with quartz, opal-CT, feldspar, muscovite, pyrite and limonite comprise the coal bearing Gondwana sequence of the study area. In Barapukuria Coal Basin the average percentage of carbon is 64.59%, hydrogen is 4.67%, nitrogen is 3.24% and sulfur is 0.45% (Pal et al. 2018).

The Gondwana Group is underlain by the Dupi Tila Formation in the studied coal Basin (Wardell 1991). The Dupi Tila Formation constitute the main aquifer of the Barapukuria Coal Mine area. The Dupi Tile sandstone is characterized by the low ZTR index (Roy et al. 2004).The Plio-Pleistocene medium to coarse-grained Dupi Tila sandstone is mainly composed of 95.17% SiO_2 and 2.11% Al_2O_3 on an average and the study of the mineralogical composition shows the presence of quartz, feldspar and lithic grains in the sandstone (Rahman et al. 2014).The geochemical study of the water of the Phulbari area reveals that the major cation facies are Ca^{2+} and Na^{+} whereas the major anion facies are HCO_3^{1-} and SO_4^{2-} (Howlader et al. 2014).

Barapukuria Coal Mine is located in Phulbari, Dinajpur, Bangladesh. Its geographical coordinates are 25˚31´45´´ N to 25˚33´05´´ N latitude and 88˚ 57´48´´E to 88˚58´53´ E longitude (Figure 1). The pH value of all water samples around the Barapukuria coal mine are above 7 which indicate the alkaline nature of the water and the colour of the coal leached drainage water is blackish and the colour of the irrigation water is slightly blackish (Fardushe et al. 2014). The present research work is carried out to show the heavy metal concentrations of the groundwater in order to determine their source and contamination level using contamination index of Backman et al., 1998.

Fig. 1: Map of the Study Area.

2. Methodology

Collection of Water Samples
Sixteen mine drainage and nearby wetlands drainage water and two groundwater samples were collected from coal mine area of Phulbari Upazilla in March, 2014. The water samples were collected in 500 ml plastic bottles, which were corked immediately to make them airtight. Samples were acidified during collection and stored in a refrigerator at 4°C. All necessary water sampling guidelines were followed during sampling. The physical parameters such as pH, EC, and temperature were measured in the field. The concentrations of major trace elements were measured by atomic absorption spectrometer (Shimadzu, AA 240FS) in the Analytical Research Division Laboratories, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka.

Methods of Water Samples Analysis
Varian spectra AA 240 FS and Graphite furnace atomic absorption spectrometry (GF AAS), Varian spectra AA 240 Z were used for water analysis. The intensity of standard solutions was measured by Atomic Absorption Spectrometer (AAS) and the respective intensity values were plotted against the mg/L values in intensity versus mg/L graph to obtain standard curve. Then the measurements of the intensity of the water samples were carried out.

The Contamination Index (C_d) of Water Samples
Quality of water takes interest of the researches and consumers (for drinking and irrigation), through which populations are exposed to harmful elements from industrial, anthropogenic and/or geological origin. Many techniques are used for assessment and visualization of hazardous defined elements. One of the approaches to calculate contamination of water bodies is by using contamination index, which takes into consideration both the number of parameters exceeding the upper permissible limits or guide values of the potentially harmful elements (Backman et al., 1998). Calculation of the contamination degree (Cd) was carried out separately for each analyzed sample of water, as a sum of the contamination factors of individual components exceeding the upper permissible values. Hence, the contamination index summarizes the combined effects of several quality parameters considered harmful to household water. According to (Backman et al., 1998), the calculation scheme of contamination index (Cd) is as follows:

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

Where,

$$C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1$$

C_{fi} = Contamination factor for the i-th component
C_{Ai} = Analytical value of the i-th component
C_{Ni} = Upper permissible concentration of the i-th component (N denotes the 'normative value')

3. Results and discussion

Geochemistry of groundwater

Table 1: Statistical Summary of Results of Chemical Analysis of Drainage Water, BCM, Dinajpur

Parameters	Fe mg/L	Mn mg/L	Zn mg/L	Ni mg/L	Cr mg/L	Pb mg/L	As mg/L	Co mg/L	Ca mg/L	K mg/L
Mean	1.987	0.303	0.072	0.068	0.019	0.004	0.000	0.002	37.712	9.405
Median	1.794	0.161	0.054	0.067	0.019	0.002	0.000	0.000	37.139	9.513
SD	0.651	0.370	0.044	0.012	0.003	0.005	0.001	0.003	7.762	3.118
Variance	0.424	0.137	0.002	0.000	0.000	0.000	0.000	0.000	60.253	9.725
Skewness	-0.047	3.701	1.222	0.232	1.024	2.891	3.013	1.584	1.031	-0.909

Kurtosis	-0.306	14.664	1.359	0.795	1.519	8.115	8.727	0.930	4.914	1.543
Max	3.079	1.766	0.190	0.095	0.027	0.020	0.005	0.008	61.470	14.450
Min	0.576	0.088	0.018	0.044	0.015	0.001	0.000	0.000	20.715	2.023
Range	2.503	1.678	0.172	0.051	0.012	0.019	0.005	0.008	40.755	12.427

Table 1 shows the chemical composition of the groundwater of Phulbari area, Dinajpur, Bangladesh. The concentration of calcium is highest on the basis of the mean content of the metals. The maximum concentration of calcium is 61.470 mg/L in Phulbari area. The calcium derived from the dissolution of carbonate rocks (Howlader et al. 2014: Moni et al. 2019). Potassium is the second abundant metal in the investigated groundwater samples which mean value is 9.405 mg/L. The enrichment of potassium in groundwater is resulted from the use of urea fertilizers (Divya and Belagali, 2012: Saha et al., 2019). The chemical weathering of sylvite and silicates especially clay minerals are the principal source of potassium in the groundwater samples of the investigated area (Sultana 2009). Table 2 shows that two of the groundwater samples exceed the permissible limit of potassium of WHO 2004.

Table 2: Correlation Between the Groundwater Samples of BCM

Param-eters	Unit	WHO drinking water stand-ard WHO (2004)	BD Guideline (2009)	Groundwater in the study area					Number of samples above acceptable limit
				Min.	Max	Aver-age	Me-dian	Stdev	
Fe	mg/L	0.3-1.0	1.0	0.576	3.079	1.987	1.794	0.651	17 Samples
Mn	mg/L	0.4	1.0	0.088	1.766	0.303	0.161	0.370	2 Samples
Zn	mg/L	3.0	5.0	0.018	0.190	0.072	0.054	0.044	None
Ni	mg/L	0.02	0.1	0.044	0.095	0.068	0.067	0.012	None
Cr	mg/L	0.05	0.05	0.015	0.027	0.019	0.019	0.003	None
Pb	mg/L	0.01	0.05	0.001	0.020	0.004	0.002	0.005	2 Samples
As	mg/L	0.01	0.05	0.000	0.005	0.0009	0.000	0.001	None
Co	mg/L	0.05	-	0.000	0.008	0.002	0.000	0.003	None
Ca	mg/L	75-200	-	20.715	61.470	37.712	37.139	7.762	None
K	mg/L	12	12	2.023	14.450	9.405	9.513	3.118	2 Samples

The mean abundance of the heavy metals of the groundwater samples can be expressed as Fe>Mn>Zn>Ni>Cr>Pb>Co>As (Figure 2). Among the heavy metals the mean concentration of iron is highest and the numerical value is 1.987 mg/L. Seventeen of the groundwater samples exceed the maximum permissible limit of iron (WHO 2004). The maximum concentrations of Mn is 1.766 mg/L and two of the samples have higher content of the permissible limit of WHO 2004. The mean concentrations of arsenic of the groundwater samples are 0.00099 mg/L. The arsenic concentrations of the main aquifer Dupi Tila Formation varies from 2.9 to 6.6 ppm (Rahman et al. 2014), which is lower than the aquifer sediments of the arsenic prone groundwater areas of Bangladesh. All of the groundwater samples have the As concentrations below the permissible limit of WHO 2004.

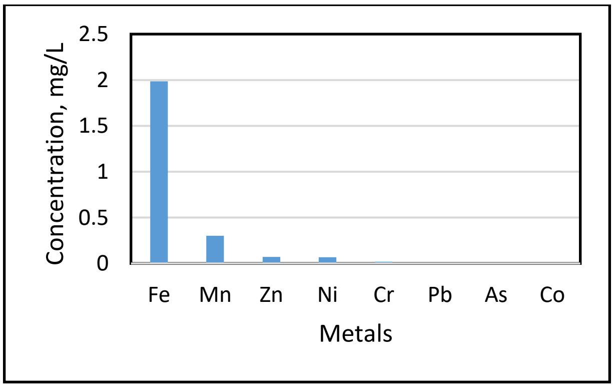

Fig. 2: The Mean Concentration of Heavy Metals in the Groundwater of Phulbari Area

The mean cobalt content of the groundwater samples of the study area is 0.002 mg/L and none of the samples exceeds the maximum permissible limit of WHO 2004. The cobalt concentration of the aquifer sediments ranges from the below detection limit to 6 ppm in the sediments of Surma Basin (Rahman et al. 2014). This may result the lower concentration of cobalt in the groundwater of Phulbari.

Table 3: Pearson Correlation Coefficient Matrix of Drainage Water of BCM

	Fe	Mn	Zn	Ni	Cr	Pb	As	Co	Ca	K
Fe	1.000									
Mn	0.359	1.000								
Zn	0.288	0.023	1.000							
Ni	0.682**	0.524*	0.242	1.000						
Cr	0.256	-0.058	-0.022	0.125	1.000					
Pb	-0.093	0.000	-0.176	-0.133	0.385	1.000				
As	0.158	-0.087	0.333	0.037	-0.361	-0.076	1.000			
Co	-0.046	0.121	0.058	0.292	-0.531*	-0.252	0.408	1.000		
Ca	0.538*	0.666**	0.295	0.644**	-0.005	-0.372	0.015	0.184	1.000	
K	0.247	-0.026	0.291	0.417	0.156	-0.292	0.051	-0.024	0.620**	1.000

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

The iron is derived from the chemical weathering of iron bearing minerals like pyrite, arsenopyrite. The arsenic shows insignificant positive correlations with iron (Figure 3) and which reveals that the arsenic is released in the groundwater by the oxidation of iron sulfide minerals (Reza et al. 2010c). The insignificant negative correlations between As and Mn is indicative of their derivation from the different sources.

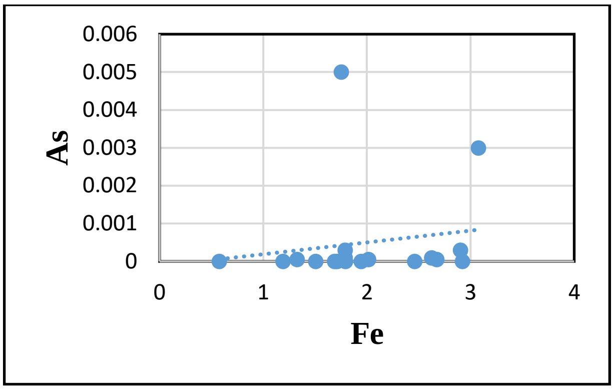

Fig 3: The Positive Correlation of Fe and As.

4. Factor analysis

From the factor analysis it can be concluded that first four factors comprise the ratios of 32.32%, 20.44%, 13.20% and 11.32% respectively and total 77.276%. The first factor (Factor -1) is characterized by the dominance of Ca, Fe and Ni, that principally indicates the dissolution of Fe and Ca bearing minerals like pyrite and carbonates. The second factor (Factor-2) is the indicator of Cr contamination within the study area. The third factor (Factor-3) is characterized by the dominance of K and Zn which reveals the influx of metal from the agricultural activities within the groundwater. Factor-4 is influenced by the fluctuations of Pb and As metals in the groundwater samples of the investigated area.

Table 4: Total Variance Explained

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	3.232	32.321	32.321	3.232	32.321	32.321
2	2.044	20.439	52.761	2.044	20.439	52.761
3	1.320	13.200	65.961	1.320	13.200	65.961
4	1.131	11.315	77.276	1.131	11.315	77.276
5	.698	6.983	84.259			
6	.597	5.974	90.233			
7	.493	4.931	95.164			
8	.287	2.867	98.031			
9	.175	1.746	99.777			
10	.022	.223	100.000			

Extraction Method: Principal Component Analysis.

Table 5: Varimax Rotated Factors for First Four Components

	Component			
	Factor -1	Factor -2	Factor -3	Factor -4
Fe	.717	.301	.034	.343
Mn	.591	.151	-.671	.055
Zn	.455	-.147	.562	.301
Ni	.843	.162	-.175	.134
Cr	-.020	.859	.224	.143
Pb	-.388	.450	-.187	.607
As	.199	-.617	.278	.561
Co	.293	-.733	-.280	.097
Ca	.907	.085	-.096	-.236
K	.592	.131	.522	-.366

Extraction Method: Principal Component Analysis.
a. 4 components extracted.

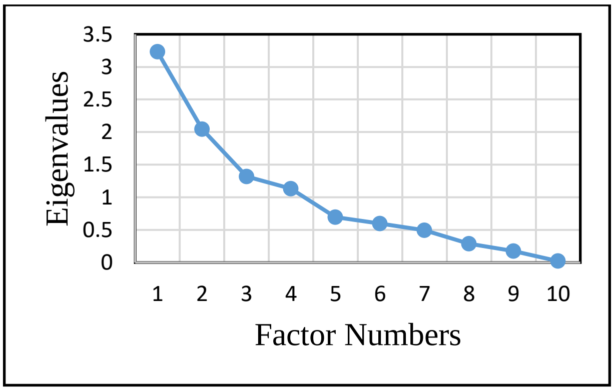

Fig. 4: Scree Plot of Eigenvalues

5. The Contamination Index (Cd) of Groundwater

Table 6 shows the contamination index of the studied groundwater samples. 88.89% of the groundwater samples show low level of contamination, 5.56% medium level of contamination and 5.56% indicative of high contamination. From table 2 it can be summarized that most of the groundwater samples are contaminated by iron and minor amounts of the groundwater samples are contaminated by the excessive amounts of Mn and K.

Table 6: Classification of Contamination Index (Backman et al. 1998)

C_d	Class	Conclusion
$C_d<1$	Low Contamination	88.89% samples
C_d 1-3	Medium Contamination	5.56% samples
$C_d>3$	High Contamination	5.56% samples
C_d = Contamination Index		

6. Conclusion

The Plio-Pleistocene Dupi Tila sandstone aquifer is composed of quartz, feldspar and lithic grains. The average amount of SiO_2 is 95.17% whereas the arsenic content varies from 2.9 to 6.6 ppm. The present research work shows that the coal mining activities have less influence on the contamination of groundwater composition of the mining area Phulbari as 88.89% of the groundwater have low contamination index (Cd). The concentration of iron is higher than the desired amount which shows the dissolution of iron bearing minerals like pyrite. The higher amount of potassium might have resulted from the agricultural activities of the investigated area. The insignificant positive correlation between Fe and As reveals that the influx of arsenic from the oxidation of iron sulfide minerals. The low concentration of arsenic in the groundwater indicates that the aquifer sediments have low arsenic content and/or fast flowing nature of the groundwater. The low cobalt content of the groundwater reflects the low cobalt concentration of the aquifer sediments. The factor analysis reveals that four principal factors constitute 77.276% of the groundwater samples and the dominance of calcium is indicative of dissolution of carbonate rocks. The further research works can be taken focusing on the physical characteristics of groundwater and mine drainage water like pH, TDS, TSS and their impact on the contamination of the soil resources of the study area.

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