Assessing the groundwater quality and health risk: A case study on Setabganj sugar mills limited, Dinajpur, Bangladesh

Syed Md. Sazzad Hossain*, Md. Emdadul Haque*, Md. Abdul Hadi Pramanik*, Md. Jalal Uddin* and Md. Abdullah Yousuf Al Harun*

*Department of Disaster Management, Begum Rokeya University, Rangpur, Bangladesh; College of Applied Meteorology, Nanjing University of Information, Science and Technology, Nanjing, China; Environmental Science Discipline, Khulna University, Khulna, Bangladesh

ABSTRACT
The poorly managed effluents from industrial activities in Bangladesh contaminate groundwater with subsequent health risks as the most of the Bangladeshi people depend on groundwater for their drinking water sources. This study aimed to investigate the contamination level in the groundwater surrounding the Setabganj Sugar Mills Limited, Dinajpur, Bangladesh with special attention on the associated health risk of the dwellers residing around it. Stratified random sampling was adopted to collect 12 water samples from tubewell to identify concentration of heavy metals using atomic absorption spectroscopy. We found metal concentration in groundwater in the order of Mn>Fe>Co>Cu>Pb>Zn>Cr among which mean value of Mn concentration exceeded the drinking water standard while Fe and Pb concentrations in few samples exceeded the standards. Metal Index, Degree of Contamination, Pollution Load Index and Heavy Metal Pollution Index confirmed more groundwater contamination at the vicinity of sugar mill compared with the other samples that taken from far away of sugar mill. Health risk assessment ensures that infants and children have a great susceptibility to the contaminated groundwater all over the study area while the adults are at high risk only in the sugar mill region. Principal Component Analysis and Factor Analysis reveal that sugar mill contributes largely to groundwater contamination along with other anthropogenic and natural sources. The findings of this study will help the environmental managers and policy makers to understand the potential health risks from the effluent of sugar mills and will knock them to treat it prior to discharge.

INTRODUCTION
About 1.1 billion (one out of six) of the total population in the world lacks access to safe drinking water despite water is a basic need (UNDP, 2015; WHO, 2017). The policy makers are busy and under pressure to achieve sustainable development goal (SDG-6) to ensure safe water for all by 2030. Unplanned urbanization, rapid industrialization, agricultural activities, and natural geochemical processes, particularly in developing countries are directly triggering pressure in this regard affecting chemical composition of groundwater day by day (Islam, Ahmed, Bodrud-Doza, & Chu, 2017b). Heavy metals intrusion in groundwater is a critical environmental problem all around the world, now a days (Kumar, Delson, & Babu, 2012). Literature suggests the release of numerous heavy metals in the atmosphere and water bodies from different industrial activities in Bangladesh (Rahman et al., 2012; Howladar et al., 2014; Khan, Seddique, Rahman, & Shimizu, 2017; Haque, Reza, & Ahmed, 2018). The arsenic contamination (BBS, 2009) and salinity intrusion (Rahman, Majumder, Rahman, & Halim, 2011) in the groundwater of Bangladesh has exacerbated the dilemma of safe drinking water throughout the country as 97% of the total population depends on groundwater for their drinking water sources (Flanagan, Johnston, & Zheng, 2012). Sugar mills have been imposing threat to the surrounding air and water environment in Bangladesh though they are providing necessity of most of the sugars in the country. Alteration of surface water chemistry from discharging the effluent of sugar mills have already been identified by the authors (Salequzzaman, Islam, Tasnuva, Kashem, & Masud, 2008).

The dwellers of Setabganj Pourosbowa (a small administrative unit known as a municipality) are experiencing water pollution and waste disposal issues including drainage congestion and bad odor from sugar mills located there. Evidence that explores intrusion of contaminants to the groundwater from the garbage and solid wastes in this area (Islam et al., 2017b) has exacerbated the nexus of industrial activities and contamination of groundwater. Further, the effluent that directly discharged from sugar industries may contaminate the adjacent groundwater aquifers through seepage and leaching. These contaminants in drinking water may result severe health problems including
hypertension, hyperkeratosis, restrictive lung disease, peripheral vascular disease, gangrene, and even cancer (Smith, Lingas, & Rahman, 2000). Hence, it is necessary to investigate the groundwater quality and health risk of individuals at the premises of sugar mills.

Previous studies have reported that heavy metals contamination in groundwater may vary region to region (e.g., Aggarwal et al., 2000; BGS and DPHE, 2001; Dowling, Poreda, Basu, Peters, & Aggarwal, 2002; Harvey et al., 2002; Islam, Shen, & Bodrud-Doza, 2017c; McArthur, Ravenscroft, Safiulla, & Thirlwall, 2001; Reza et al., 2010; Stollenwerk et al., 2007) due to varied socioeconomic activities and geo graphical locations. Numerous studies have been conducted to identify the contribution of industrial activities to surface and groundwater contamination in Bangladesh. However, none of them have focused on the contribution of sugar mills to heavy metal contamination in groundwater despite the effluent of sugar mills contains substantial concentration of heavy metals. To our knowledge, there is no study that analyzed the health risk of the residents around sugar mills though very few studies covered about the other industries. Therefore, the current study investigated the groundwater quality at the vicinity of the sugar mill in Setabganj Pouroshova with particular attention on health risk assessment of the nearby dwellers. Thus, our research questions stand as: (i) what is the state of the heavy metal concentrations in the groundwater of the study area? (ii) Is there any potential health risk to different aged people who drink contaminated groundwater? And (iii) what are the probable sources of groundwater pollutants? Concerning these, the specific objectives of the study are: (a) to assess the groundwater quality of the study area, (b) to assess the associated health risk for consuming contaminated drinking water and (c) to identify probable sources of the groundwater pollutants.

Materials and methods

General description of the study area

The study was conducted to assess the groundwater quality surrounding the Setabganj Sugar Mills region at Setabganj Pouroshova (small city) in Dinajpur District, Bangladesh (Figure 1). The study area is situated between 25.82° N to 25.75° N latitude and 88.45°E to 88.49° E longitudes. This Pouroshova consists of eight wards (smallest administrative unit) and the sugar mill is located at ward no. 8.

Water sample collection and preservation

Stratified random sampling technique was adopted to collect eight groundwater samples from the local tube wells with the consideration of one sample from each ward. However, extra three samples were collected from ward no. 8 (where the sugar mill is situated) to gain a deep insight about the impact of the sugar mill. Prior to selection of the tube wells the depth record was collected from the owners of the wells to confirm similar depth tube wells in this study. Table 1 provides a detail information about the sample and tube well depth information. Each tube well was run for five minutes prior to sample collection to ensure that sample water was coming from the aquifer, not from the pipe. The groundwater samples

Figure 1. Location map of the study area illustrating the sample points.
were collected in cleansed high-density polypropylene (HDPP) bottles. A mixture of 1:3 concentrated HCl and HNO₃ was added with the samples to preserve in room temperature at the chemistry laboratory of Begum Rokeya University, Rangpur as soon as they were collected. Samples were then taken to the laboratory for laboratory tests within 3 days.

Quantifying of heavy metals concentrations in laboratory analysis

Atomic absorption spectroscopy (AAS) is generally used to quantify the concentration of heavy metals in water utilizing the absorption of optical radiation by gas atoms. A group of Australian scientists led by Sir Alan Walsh at the Commonwealth Scientific and Industrial Research Organization (CSIRO) modified the AAS to the currently used form during 1950s (Koirtyohann, 1991). The water samples were digested in the chemistry laboratory of Begum Rokeya University, Rangpur to get ready for AAS. Then the samples were taken to the chemistry laboratory of Rajshahi University to quantify heavy metals concentration using flame AAS method.

Groundwater quality assessment using metal index (MI)

The metal index (MI) was preliminarily defined by Tamasi and Cini (2004). A series of studies reported MI as a quick tool for assessing the overall quality of drinking water (Bakan, Ozkoc, Tulek, & Cuce, 2010; Caeiro et al., 2005; Tamasi & Cini, 2004). Equation one reflect the MI.

\[ Ni = 1MI = \sum C_i / MAC \]  \hspace{1cm} (1)

Where MI is the metal index, C is the concentration of each element in the solution, MAC is the maximum allowed concentration for each element and the subscript \( i \) is the \( i \)th sample. The threshold level is MI<1. Table 2 represents MI water quality classifications.

Groundwater quality assessment using degree of contamination \( (C_d) \)

The Degree of Contamination \( (C_d) \) is another index that uses the contamination factor to identify the level of contamination. The index can be calculated by the following equation 2 (Backman, Bodii, Lahermo, Rapant, & Tarvainen, 1998).

\[ C_d = \sum_{i=1}^{N} C_{fi} \]  \hspace{1cm} (2)

Here, \( C_{fi} \) is the contamination factor, which is calculated by Equation 3

\[ C_{fi} = (C_{ai}/C_{mi}) - 1 \]  \hspace{1cm} (3)

Here, \( C_{ai} \) is the analytical value of the \( i \)th variable and \( C_{mi} \) is the upper permissible concentration (MAC = Maximum allowed concentration). MAC values were taken from Bangladesh Standard (1997). The \( C_d \) index classification is shown in Table 3.

Groundwater quality assessment using pollution load index (PLI)

The Pollution Load Index (PLI) is obtained as Contamination Factors \( (C_f) \). This \( C_f \) is the quotient obtained by dividing the concentration of each metal by maximum allowed concentration (MAC). MAC values were taken from Bangladesh Standard. The PLI was used in a number of literatures with slight modification (Soares, Boaventura, & Esteves da Silva, 1999) indicating the value of >1 is polluted and <1 is not polluted (Harikumar, Nasir, & Rahma, 2009; Thambavani & Mageswari, 2013). Originally, pollution load index was developed by Tomlinson, Wilson, Harris, and Jeffrey (1980), as placed in equation 4 and 5.

\[ C_f = C_{metal}/C_{MAC} \]  \hspace{1cm} (4)

\[ PLI = \sqrt{[n]C_{f1} \times C_{f2} \times C_{f3} \times C_{fn}} \]  \hspace{1cm} (5)

| Class | MI value | Water quality |
|-------|----------|---------------|
| I     | <0.3     | Very pure     |
| II    | 0.3-1    | Pure          |
| III   | 1-2      | Slightly affected |
| IV    | 2-4      | Moderately affected |
| V     | 4-6      | Strongly affected |
| VI    | >6       | Seriously affected |

Table 2. MI water quality classification (Caeiro et al., 2005).

Table 1. Groundwater samples depth and respective ward no. and coordinates.

| Sample no. | Ward no. | Depth (meter) | Latitude (decimal degree) | Longitude (decimal degree) |
|------------|----------|---------------|---------------------------|---------------------------|
| 1          | 2        | 300           | 25.8082                   | 88.4540                   |
| 2          | 65       | 25.8118       | 88.4616                   |                           |
| 3          | 35       | 25.8081       | 88.4615                   |                           |
| 4          | 40       | 25.8052       | 88.4601                   |                           |
| 5          | 50       | 25.8057       | 88.4647                   |                           |
| 6          | 40       | 25.7988       | 88.4632                   |                           |
| 7          | 40       | 25.7948       | 88.4643                   |                           |
| 8          | 45       | 25.7931       | 88.4516                   |                           |
| 9          | 60       | 25.7974       | 88.4544                   |                           |
| 10         | 60       | 25.7976       | 88.4546                   |                           |
| 11         | 60       | 25.7982       | 88.4564                   |                           |
| 12         | 60       | 25.7968       | 88.4583                   |                           |
Where, \( C_f \) = contamination factor, \( n \) = number of metals, \( C_{metal} \) = metal concentration in polluted waters, \( C_{MAC} \) = maximum allowed concentration value of that metal.

**Groundwater quality assessment using heavy metal pollution index (HPI)**

HPI calculates the total groundwater quality in relation with heavy metals. HPI calculation is conducted by appointing a weightage \( (W_i) \) for every metal. The weightage is an arbitrary value between zero and one, reflecting the relative importance of individual quality considerations, and can be defined as inversely proportional to the recommended standard \( (S_i) \) for each parameter (Horton, 1965; Mohan, Nithila, & Reddy, 1996). The highest tolerant value for drinking water \( (S_i) \) refers to the maximum allowable concentration in drinking water in absence of an alternate water source. The desirable maximum value \( (I_i) \) indicates the standard limits for the same parameters in drinking water. The HPI model given by Mohan et al. (1996) is presented in equation 6.

\[
HPI = \sum_{i=1}^{n} W_i Q_i \sum_{i=1}^{n} W_i
\]

(6)

Where \( Q_i \) is the subindex of the \( i^{th} \) parameter, \( W_i \) is the unit weightage of the \( i^{th} \) parameter and \( n \) is the number of parameters considered. The subindex \( (Q_i) \) of the parameter is calculated by equation 7.

\[
Q_i = \sum_{i=1}^{n} \frac{|M_i - I_i|}{S_i - I_i}
\]

(7)

Where \( M_i \) is the monitored value of the heavy metal of \( i^{th} \) parameter, \( I_i \) is the ideal value of the \( i^{th} \) parameter, and \( S_i \) is the standard value of the \( i^{th} \) parameter in ppb. The quantity \( |M_i - I_i| \) indicates a numerical difference between the two values, ignoring the algebraic sign; that is the absolute value. Generally, the critical pollution index of HPI value for drinking water is 100 (Prasad & Bose, 2001). On the other hand, Vetrimurugan, Brindhna, Elango, and Nd wandwe (2017) used following classification i.e., excellent \( (0-25) \), good \( (26-50) \), poor \( (51-75) \), very poor \( (76-100) \), and unsuitable \( (100) \). In computing the HPI, Prasad and Bose (2001) measured \( W_i \), as \( 1/MAC \) (maximum admissible concentration) of the corresponding metal as proposed by Siegel (2002). This approach has been applied in this work.

**Health risk assessment using hazard quotient (HQ) and hazard index (HI)**

Risk of health hazard (noncarcinogenic risk or chemical toxicity) related to the direct ingestion of contaminated water through drinking can be calculated using equation 8 provided by USEPA (1989).

\[
LADD = (C*IR*ED*EFBW*AT)
\]

(8)

Where, \( LADD \) = Lifetime average daily dose of ingestion through drinking water \( (mg/kg/day) \), \( C = \) Concentration of metal \( (mg/L) \), \( IR = \) Ingestion rate of water \( (250 \text{ mL/day for infants, } 1.5 \text{ L/day for children, } 3 \text{ L/day for adults}) \), \( EF = \) Frequency of exposure \( (\text{days/year}) \), \( ED = \) Duration of exposure/average-year, \( BW = \) Body weight \( (6.9 \text{ kg for infants, } 18.7 \text{ kg for children and } 57.5 \text{ kg for adults}) \), and \( AT = \) Average time \( (\text{days}) \) (Vetrimurugan et al., 2017).

It is assumed that water is consumed throughout the year (exposure frequency) in a lifetime (exposure duration) by an individual. In this case, the average time will be equal to exposure frequency and exposure duration. Thus the equation can be simplified as follows (Equation 9):

\[
LADD = (C*IRBW)
\]

(9)

Hazard quotient and Hazard Index was calculated following US EPA (1989) as given in Equation (10) and (11).

\[
HQ = \frac{LADD*RFD}{D}
\]

(10)

\[
HI = \sum_{i=1}^{N} HQ_i
\]

(11)

Where RFD is the reference dose of heavy metal that an individual can be exposed (Table 4).

**Potential source apportionment using multivariate statistics**

Multivariate statistical approaches are very useful in attaining significant information from the hydro-chemical dataset in the groundwater system. In this study, the multivariate statistical approaches including principal component analysis (PCA), factor analysis (FA), and cluster analysis (CA) were successfully applied to analyze the hydro-chemical data of groundwater. In order to understand the geochemical processes and the sources of the heavy metals in the groundwater, hydro-chemical data were subjected to PCA that allowed to grouping them based on their inherited properties. The PCA was performed with an

| Tables | Values |
|--------|--------|
| Metals | RFD    |
| Chromium | \( 3 \times 10^{-3} \) |
| Copper | \( 5 \times 10^{-3} \) |
| Lead | \( 3.6 \times 10^{-3} \) |
| Manganese | \( 1.4 \times 10^{-3} \) |
| Zinc | \( 3 \times 10^{-3} \) |
| Iron | Not available |
| Cobalt | Not available |
orthogonal Kaiser’s Varimax rotation to make the factors more interpretable without changing the original mathematical dataset. The varimax rotation can successfully reduce the contribution of less significant parameters in the groundwater quality obtained from the PCA. The first PCA was accounting for the highest variance in the dataset followed by the next PCA and so on. Additionally, the cluster analysis (CA) was employed to identify similar groups or clusters based on similar characteristics within the class and dissimilar characteristics among various classes. The results of attaining multivariate statistical methods were assessed by the component plot, scree plot and dendrogram based on Ward’s method. All the statistical analyzes were performed by using the SPSS software version 22.0.

**General statistical techniques and spatial analysis**

Descriptive statistical methods were applied to interpret the statistical variables (maximum, minimum, mean and standard deviation) for groundwater quality data set. The correlation matrix (CM) analysis was carried out to define the degree of pair with two parameters in this study. The terms “significant/strong,” “moderate,” and “nonsignificant” are applied to Pearson’s correlation matrix analysis based on the approach proposed by Liu, Lin, and Kuo (2003) and it indicates to the values as 0.75, 0.75–0.50, and 0.50--0.30, respectively.

Currently, various spatial interpolation techniques such as kriging method, inverse distance weighted method, etc. are widely used for predicting and measuring the spatial variability of the groundwater data set. The inverse distance weighted (IDW) method was applied for the spatial analysis in this study because of its accuracy compared to other interpolation methods. This method is in-built within ArcGIS.

**Results and discussion**

**Concentration of heavy metal in groundwater**

Metal pollution in groundwater is governed by the several aspects including the level of weathering of the different rock, quality of the aquifer and effect of the peripheral pollution sources that ultimately, create complex groundwater chemistry (Aksever, Davraz, & Bal, 2016). It is important to consider the fact that trace level of concentration of heavy metal is needed for the functioning of metabolic actions of the human physique (WHO, 1996), however, exceeding the standard might impose severe health hazards.

Table 5 represents the concentrations of heavy metals in groundwater in the study area. The values are also compared with Bangladesh (DoE, 1997), Indian (Indian Standard, 2012), and WHO (WHO, 2011) standards; however, no standard was found for Cobalt. The mean concentration of the Mn, Fe, Co, Cu, Pb, Zn, and Cr was .0069, .2482, .0375, .0234, .0156, and .0106, respectively. All the metals were found in acceptable limits except for Mn, Fe, and Pb. The mean value of Mn concentration was exceeded all the standards. The level of Fe and Pb in few samples (ward 8) was exceeded all standards while their mean value was exceeded the Indian standard only. The standard deviation shows very less fluctuation indicating small variations in the parameters among the samples.

| Metals (mg/L) | Min  | Max  | Mean  | Std. deviation | Variance | Bangladesh standard (1997) | WHO limits (2011) | Indian Standard (2012) acceptable limits | Indian Standard (2012) permissible limits |
|---------------|------|------|-------|----------------|----------|--------------------------|-------------------|------------------------------------------|------------------------------------------|
| Co            | .0070| .0405| .2482 | .1102          | .012     | -                        | 2                 | 5                                        | 15                                        |
| Zn            | .0021| .0084| .0156 | .0321          | .001     | 5                        | 1                 | 0.3                                      | No relaxation                             |
| Fe            | .0964| .7352| .3452 | .1894          | .036     | 0.3- 1.0                 | 1                 | 0.3                                      | No relaxation                             |
| Cu            | .0139| .0551| .0375 | .0136          | .000     | 1.00                     | 2.00              | 0.05                                     | 1.5                                      |
| Cr            | .0026| .0230| .0106 | .0064          | .000     | 0.05                     | 0.05              | 0.05                                     | No relaxation                             |
| Pb            | .0000| .0720| .0234 | .0187          | .000     | 0.05                     | 0.01              | 0.01                                     | No relaxation                             |
| Mn            | .0494| .1661| .4069 | .4973          | .247     | 0.1                      | 0.1               | 0.1                                      | 0.3                                      |
found substantial heavy metal concentration in the groundwater of industrial zone (Rahman et al., 2012), mining field (Howladar et al., 2014; Khan et al., 2017), and agricultural field (Brevik & Burgess, 2012) in Bangladesh that support the findings of our study.

**Groundwater quality evaluation using MI**

Metal Index is a general index for assessing the groundwater quality which defines the concentration of metals in groundwater in relation to maximum allowable concentration (MAC). In this study, the MAC values have been taken from the Bangladesh standards (Bangladesh Standard (DoE), 1997). Water quality has been classified into six classes from “very pure” to “seriously affected” (Caetano et al., 2005). This study has followed the similar method used by Balakrishnan and Ramu (2016) to calculate MI. Metal indexing is one of the best water quality assessing method of heavy metal contamination as the value of the index is not affected by the number of heavy metals considered to calculate the MI. Figure 2 depicts the spatial variation of MI values in the study area map. The MI calculation reflects that half of the samples fall into the second class of the MI class which denotes pure water quality. Sample 3 and sample 9 are fallen under very pure water. Sample 11 that collected from sugar mills region found to be in the fourth class denoting moderately affected water quality. On the other hand, sample number 5, 6, and 12 that were collected from ward nos. 5, 6, and 8 (Sugar mills area) respectively reflect the worst water quality among all the samples analyzed. According to the MI class the water quality of the collected water samples is slightly affected by heavy metals. The findings of Balakrishnan and Ramu (2016) who assessed MI value with the same technique is very much consistent with this study as the water quality in their study was also slightly affected by heavy metals.

**Groundwater quality evaluation using C_d**

Degree of Contamination is an index to assess heavy metal contamination in groundwater. The C_d was evaluated individually for each water sample. The sum of the contamination factors of individuals crossed the maximum allowable value. The C_d reflects the accumulated effects of each harmful metal. C_d is the most precise way in evaluating water quality as it considers all the heavy metals in calculation. Thus, to get more precise result using C_d all the heavy metals concentration needs to be quantified. However, for this study, six heavy metals have been taken into consideration in computing C_d. Edet and Offiong (2002) categorized the values of C_d into three categories, where C_d<10 means low pollution, 10<C_d<20 means medium pollution and C_d exceeding 20 is high pollution in water bodies.

Figure 3 depicts the spatial distribution of the computed C_d value for the study area. The figure shows that sample 11 and 12 that were collected from the sugar mill region have the highest C_d values among all the samples. These two water samples fall under “Medium Pollution” class while rest of the samples had low pollution level according to the C_d calculation and classification (Edet & Offiong, 2002). Bodrud-Doza et al. (2016) in their study of central Bangladesh revealed that the ranges and mean value of C_d of the groundwater samples were 0.19–32.63 and 7.51, respectively.

**Groundwater quality evaluation using PLI**

The limitation of C_d (not considering all the metals) may be overcome in PLI to assess the metal accumulation pollution in groundwater. The PLI value of >1 is polluted whereas <1 indicates no pollution (Thambavani & Mageswari, 2013). All the samples in this study have been found <1 in PLI index which indicates that the water is not polluted (Table 6). In contrast, Likuku, Mmolawa, and Gaboutloeloe (2013)

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Table 6. Comparison of values of different metal contamination assessing indices.

| Sample | MI value | Water quality | C_d  | Pollution | PLI | Polluted | HPI |
|--------|----------|---------------|------|-----------|-----|----------|-----|
| 1      | 0.4237   | Pure          | 2.5422 | Low       | 0.0982 | No       | 11,1490 |
| 2      | 0.4008   | Pure          | 2.4048 | Low       | 0.1258 | No       | 6.6196  |
| 3      | 0.467    | Very Pure     | 1.4805 | Low       | 0.0682 | No       | 16,6412 |
| 4      | 0.4820   | Pure          | 2.8921 | Low       | 0.1130 | No       | 62,1648 |
| 5      | 1.1954   | Slightly affected | 7.1726 | Low       | 0.1835 | No       | 11,2646 |
| 6      | 1.0042   | Slightly affected | 6.0256 | Low       | 0.1557 | No       | 28,6836 |
| 7      | 0.3605   | Pure          | 2.1635 | Low       | 0.0826 | No       | 2,94291 |
| 8      | 0.3571   | Pure          | 2.1427 | Low       | 0.0833 | No       | 20,0646 |
| 9      | 0.2231   | Very Pure     | 1.3389 | Low       | 0.0731 | No       | 1,7287  |
| 10     | 0.6744   | Pure          | 4.0467 | Low       | 0.1131 | No       | 2,5571  |
| 11     | 2.9498   | Moderately affected | 17,6993 | Medium     | 0.1570 | No       | 11,8687 |
| 12     | 1.9554   | Slightly affected | 11,7326 | Medium     | 0.2026 | No       | 8,5630  |
| Min    | 0.2231   | 1.3389        | 0.0682 | No       | 0.6196  |
| Max    | 2.9498   | 17,6993       | 0.2026 | 62,1648  |
| Mean   | 0.8561   | 5.1368        | 0.1213 | No       | 14,8456 (not critical) |
found PLI value of 1.63 indicating polluted water while studied heavy metal enrichment around copper-nickel mine in water at Eastern Botswana.

**Groundwater quality evaluation using HPI**

Heavy metal Pollution Index is a popular indexing technique to assess the groundwater quality as it is calculated considering the maximum allowable concentration, standard permissible values and highest permissible values with the monitored concentration of heavy metals. This study calculated both the HPI values for each sample (Table 6) and the HPI value for the study area (Table 7). The overall HPI value of the study area and the average HPI value of the sample-wise calculation is 14.05684 and 14.84569, respectively. The approximately same HPI values indicate that the calculations in either way can be reliable. Most of the research studies compare their HPI values with the critical HPI value of 100. The water of HPI>100 is thought to be harmful (Balakrishnan & Ramu, 2016; Singh & Kamal, 2017; Yankey et al., 2013). On the other hand, Vetrimurugan et al. (2017) used the classification as: excellent (0–25), good (26–50), poor (51–75), very poor (76–100), and unsuitable (100). According to Vetrimurugan et al. (2017), all the samples in this study fall under excellent quality with the exception of sample number 6 (good) and 4 (poor).

Yankey et al. (2013) found the HPI of the groundwater below the critical value of 100 with the exception of one sampling point that has an overcritical HPI value of 102.97. Literatures also identified varied HPI values of 1.5 in the monsoon season and 2.1 in the postmonsoon season although, both are well below the critical index limit of 100 (Singh & Kamal, 2017).

**Health risk assessment using the HI**

Earlier in this study, the suitability of drinking water was attempted to illustrate by comparing the mean concentrations of metals with the Bangladeshtandard (1997), Indian Standard (2012) and WHO standard (2011). However, sometimes this technique alone cannot be able to establish the groundwater quality for drinking purpose as it is important to consider the volume of water a person takes in a certain period of time (Vetrimurugan et al., 2017). The HI takes into account both the quantity of water consumed by a person and the time period. In extreme scenarios, the exposure pathway may also be calculated considering that groundwater is used by the entire population for drinking purposes.

The noncarcinogenic risk given by HI was calculated for five heavy metals in this study as the RfD values for Fe and Co were not available. HI<1 is considered as safe and HI>1 presents a risk that is applicable for a single heavy metal or for many metals as a whole (Vetrimurugan et al., 2017). Table 8 reflects the computed HI values of the study area for infants, children and adults. The highest HI values for all three ages of consideration has found in the sugar mill region indicating that the drinking

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**Table 7.** Calculation of HPI of the study area taking mean concentrations as monitored (M) value.

| Heavy metals | M (µg/L) | *MAC (µg/L) | \( W_i=M_i \) | \( **S_i (µg/L) \) | \( **I_i (µg/L) \) | \( Q_i = \sum_1^{n} \left[ \frac{M_i-I_i}{S_i} \right] \) | \( \sum W_i = 0.7128 \) | \( W_i / Q_i = 10.02 \) |
|--------------|----------|-------------|----------------|-----------------|----------------|----------------------------------|-----------------|-----------------|
| Zn           | 15.675   | 5000        | 0.0002         | 500             | 3000          | 149.2163                           | 0.029843        |                 |
| Fe           | 345.275  | 2000        | 0.005          | 300             | 2000          | 145.275                            | 0.726375        |                 |
| Cu           | 37.592   | 1000        | 0.001          | 1000            | 2000          | -196.241                           | -0.19624        |                 |
| Cr           | 10.667   | 500        | 0.02           | 50              | 50            | 0                                | 0               |                 |
| Pb           | 23.4     | 500        | 0.66667        | 100             | 10           | 14.88889                           | 9.25926         |                 |
| Mn           | 406.95   | 500        | 0.02           | 100             | 500           | -23.2625                           | -0.46525        |                 |

\( \sum W_i = 0.7128 \)

\( HPI = \frac{\sum W_i}{Q_i} = \frac{14.05684}{15.9127} \)

*MAC= Maximum Allowable Concentration adapted from (Siegel, 2002 and Yankey et al., 2013), **S= Standard Permissible value, **I= Highest Permissible/ Ideal value (Yankey et al., 2013).
water quality in the sugar mill region is not safe for any age group of people.

Figure 4 illustrates the HI values for infants ensuring that the drinking water quality of the whole study area have a potential high risk for infants. So, the nexus of water contamination and infants’ health is horrible in the study area as the infants are very susceptible to a small contamination, even. Sugar mill region (ward no. 8) has the highest HI value exceeding the threshold level by a huge margin (HI>3.5). Figure 5 demonstrates that the HI value is suitable for the children only in ward no. 1 and ward no. 2 while water in ward no. 8 has the worst quality. Finally, Figure 6 depicts that the HI value in most cases was suitable for the adult drinking as they have more tolerance compared with other age group. However, the highest HI values are again found in the sugar mill region that ultimately makes this region highly risk for all age’s people in terms of HI based on drinking water quality.

Maigari, Ekanem, Garba, Harami, and Akan (2016), in their study, evaluated the health hazard for contaminated drinking water by using HQ and HI. The HQ value for Fe, Mn, Ni, and Co in parts of their study area were greater than the standard that gestures potential health risk for both adults and children while cobalt was the only heavy metal of concern in water of the other parts of the study area. However, they found that the HI values are of high risk for all the sampling sites. The non-carcinogenic risk due to ingestion of groundwater through drinking was very high and ranked as infants > children > adults in the study of Vetrimurugan et al. (2017). They also demonstrated that silver, lead, nickel, cadmium, and manganese were contributed substantially to the health hazard. Islam, Shen, and Bodrud-Doza (2017a) computed the health risk of contaminated water in Chapai-Nawabganj district, one of the most arsenic contaminated areas in Bangladesh. In their study, the cancer risk of arsenic through oral pathway was calculated 0.007 and 0.003 for children and adults respectively. On the other hand, the noncancerogenic (oral) risk was 14.70 and 6.9 for children and adults respectively, indicating serious health risks.

Table 8. Hazard Index values of each sample for infants, children, and adults.

| Sample | HI<sub>Infants</sub> | HI<sub>Children</sub> | HI<sub>Adults</sub> |
|--------|---------------------|----------------------|---------------------|
| 1      | 1.2285              | 0.8706               | 0.5662              |
| 2      | 2.8561              | 0.8468               | 0.5508              |
| 3      | 4.7550              | 1.4235               | 0.9259              |
| 4      | 4.3901              | 2.4649               | 1.6032              |
| 5      | 2.9453              | 1.0443               | 0.6792              |
| 6      | 6.8884              | 2.0764               | 1.3506              |
| 7      | 2.1630              | 1.1614               | 0.7554              |
| 8      | 1.5026              | 1.4774               | 0.9609              |
| 9      | 4.7662              | 1.4482               | 0.9419              |
| 10     | 6.5072              | 1.6753               | 1.0897              |
| 11     | 3.6817              | 2.6746               | 1.7396              |
| 12     | 8.5635              | 2.6005               | 1.6914              |
| Min    | 1.2285              | 0.8468               | 0.5508              |
| Max    | 8.5635              | 2.6746               | 1.7396              |
| Mean   | 4.1706              | 1.6470               | 1.0712              |

Figure 4. Spatial distribution map of Hazard Index (HI) values for infants.

Figure 5. Spatial distribution map of the Hazard Index (HI) values for children.
**Relationship between parameters of groundwater of the study**

Pearson’s correlation matrix was applied to find out the relationship between metals and several indices that have been used in this study earlier. The correlation matrix is a widely used method to find out the influence of parameter pairs on groundwater quality (Islam et al., 2017b). Table 9 depicts correlation coefficient matrix of heavy metals, indices and other parameters of the study. Several pairs have a very strong relationship with the very high level of significance. Manganese shows very strong relationship (99% significant) with Metal Index (r = .996), Degree of Contamination (r = .996), and PLI (r = .746) which indicates that Mn has the most impact on the groundwater quality evaluation indices in the study area.

Heavy metal Pollution Index showed very significant (99%) strong relationship with Pb. The strong positive r-value (0.926) of HPI-Pb pair explains that the value of HPI in the study area is highly dependent on Pb concentrations despite Pb concentrations were found in a lesser quantity (Table 9) and did not exceed the standards of the drinking water. The effect of Cr was found to be the most severe on the infants. Hazard Index for infants (HIi) was in a strong relationship (r = 0.994, 99% significant) with Cr. The Cu, Pb, and Mn have shown statistically significant (95%) relationship with Hazard Index for both children and adults.

Apart from the pairs of metal and groundwater quality indices, there have strong relationships of 95–99% significance between few of the indices, e.g., pairs of HI for adults (HIa) and HI for children (HIc) (r = 1), HIa – HIc(r = 0.633), HIa-HIc(r = 0.633), HIa-Cd

**Table 9. Correlation matrix of indices and parameters of the study.**

|     | Co | Zn | Fe  | Cu  | Cr  | Pb  | Mn  | MI  | Cd  | PLI | HPI | HIa | HIc | HIa | HIc | HIa-Cd |
|-----|----|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|--------|
| Co  | 1  |    |     |     |     |     |     |     |     |     |     |     |     |     |     |        |
| Zn  | .35| 1  |     |     |     |     |     |     |     |     |     |     |     |     |     |        |
| Fe  | -.02|.26| 1   |     |     |     |     |     |     |     |     |     |     |     |     |        |
| Cu  | -.50|.36|.12| 1   |     |     |     |     |     |     |     |     |     |     |     |     |        |
| Cr  | .02|.05|.46| 1   |     |     |     |     |     |     |     |     |     |     |     |     |        |
| Pb  | -.26|.32|.04| .02| .03| 1   |     |     |     |     |     |     |     |     |     |     |        |
| Mn  | -.08|.03|.14| .51| .24| -.07| 1   |     |     |     |     |     |     |     |     |     |        |
| MI  | -.10|.06|.11| .51| .26| .00| .99| 1   |     |     |     |     |     |     |     |     |        |
| Cd  | -.10|.06|.11| .51| .26| .00| .99| 1   |     |     |     |     |     |     |     |     |        |
| PLI | .02|.21|.14| .44| -.10| .74| .75| .75| 1    |     |     |     |     |     |     |     |        |
| HPI | .25|.30|.14| .92| -.13| .05| -.05| .02| 1    |     |     |     |     |     |     |     |        |
| HIa | -.02|.09|.46| .51| .99| .11| .31| .32| .32| .46| .46| .06| 1   |     |     |     |        |
| HIc | -.36|.34|.20| .65| .54| .59| .61| .67| .67| .45| .47| .63| 1   |     |     |     |        |
| HIa-Cd| .35|.12|.11| .49| -.36| -.10| -.09| -.10| -.10| -.11| -.14| -.38| -.36| -.36|    |        |

*aCorrelation is 99% significant (two-tailed).
*bCorrelation is 95% significant (two-tailed).

HIa=HI for Adults, HIc=HI for Children, HIa=HI for Adults, D = Depth.

Figure 6. Spatial distribution map of the Hazard Index (HI) values for adults.
(r = 0.671), Hf\textsuperscript{2-}C\textsubscript{d} (r = 0.671), HI\textsuperscript{2}-MI (r = 0.671), and HI\textsuperscript{1}-MI (r = 0.671).

No significant correlation has been found between the metals, in particular. Some positive and negative relationships have been observed among them but that were negligible. The phenomenon indicates that the sources of these parameters are independent to each other as described in other study (Kamrani, Rezaei, Amiri, & Saberinasr, 2016). The sources of the metals may be both natural and anthropogenic. Another fact to note that there exist negative relationships between metals and depth indicating higher concentrations of metals with decreasing depths of the wells.

The comparison of the indices is important along with the metals as each index is different from each other. The $C_d$ index is better for single parameter pollution index (Backman et al., 1998) while MI calculates the averages of the contamination factors (Bakan et al., 2010) and PLI identifies the numerical root of the contamination factor (Harikumar et al., 2009; Thambavani & Mageswari, 2013). Literature further denoted that HPI is the best among all the indices as it considers both the upper permissible value and the lower permissible value and overcomes the problem of not quantifying all the available metals in a sample (Vetrimurugan et al., 2017). Results of HPI in this current study also explain the groundwater quality in the best way compared with the other indices.

**Potential source apportionment of groundwater contaminants**

The risk of consuming contaminated groundwater can be reduced substantially if the sources of contamination are well identified. With a view to elucidating the factors influencing the parameters which affect the groundwater quality, the FA and CA was applied in a study (Islam et al., 2017a). PCA helps to group the data based on their inherent characteristics (Islam et al., 2017b). The PCA is performed with Kaiser’s varimax rotation to make the factors more interpretable without changing the original data structure. Varimax rotation is generally used to maximize the total variance of the factor coefficients explaining the possible sources that influence groundwater aquifer.

PCA can reduce the less important datasets that helps to understand which sources are more potential in creating pollution. The scree plot (Figure 7(a)) of the eigenvalues identifies components or factors to retain for clustering. Figure 7(b) is the component plot in rotated space of those retained components. Three components are retained with the Kaiser’s varimax rotation which has eigenvalues greater than 1 and that has been presented in Table 10. Table 10 represents three components with Kaiser’s varimax rotation along with eigenvalues, % of variances and cumulative % of the eigenvalues of each component. The component factors are placed against each metal in each factors eigenvalues. The first three factors compiled 71.241% of total variances.

The first component elucidates that 31.439% of total variances is positively loaded with Zn, Co, and Mn (geogenic and anthropogenic sources). The Mn might be originated by geogenic sources, which could be released by chemical weathering of parent materials and geochemical alteration of carbonate minerals (Bodrud-Doza et al., 2016). Potential heavy metals like Zn can be assimilated in groundwater through leaching of metals from industrial activities. Consistently, in our study several industries (mainly sugar mill) are located in the study area that might be responsible for heavy metal contamination in the studied groundwater.

Component 2 comprises 24.687% of total variance and is positively loaded with Mn, Cu and Cr indicating that the sources of these three metals are same. This

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**Figure 7.** Principal component analysis by a) scree plot of the eigenvalues, and b) component plot in rotated space.
Table 10. Varimax rotated component matrix.

| Parameters | Components 1 | Components 2 | Components 3 |
|------------|--------------|--------------|--------------|
| Zn         | .770         | -.137        | .182         |
| Co         | .716         | -.255        | .042         |
| Pb         | -.704        | -.460        | .192         |
| Mn         | .006         | .782         | .152         |
| Cu         | -.455        | .780         | .238         |
| Fe         | -.180        | -.024        | -.860        |
| Cr         | -.095        | .289         | .803         |
| Eigenvalues| 2.201        | 1.728        | 1.058        |
| % of Variance | 31.439 | 24.687 | 15.115 |
| Cumulative % | 31.439 | 56.126 | 71.241 |

Extraction Method: Principal Component Analysis.
Rotation Method: Varimax with Kaiser Normalization.

Component 3 comprises 15.15% of total variances and positively loaded with each of the studied metals except Fe which explains that Mn, Co, Cu, Pb, Zn, and Cr are originated from the same source while the sources of Fe is varied. Natural sources like oxidation of iron (Rahman & Gagnon, 2014) and rainwater through the leaching of secondary salts infiltrated into aquifer may be an option for a possible source of groundwater contamination with Fe (Bodrud-Doza et al., 2016).

Hierarchical CA was also employed to interpret the relationship of probable sources of pollutants in the groundwater that is presented in a dendrogram (Figure 8).

Three clusters were retained from the dendrogram. Cluster 1 retains Cr, Pb, Cu, and Zn indicating mixed but same sources. Probable origin may be natural hydrogeological, leaching, municipal and industrial wastes. Cluster 2 retains Co and Fe indicating different sources from the cluster 1. On the other hand, Mn is independently originated from different sources.

Conclusions

Findings of this study reveal that the studied groundwater samples are contaminated with Mn, Fe, and Pb. Though Co, Cu, Zn, and Cr are also existed in the water samples but they did not exceed the standard. The mean values of the concentration of identified heavy metals are found in the order of Mn>Fe>Co>Cu>Pb>Zn>Cr. The analysis of anthropogenic and natural sources of heavy metals contamination in the groundwater through PCA, FA, and CA techniques confirm the contribution of sugar mill to the heavy metal contamination in the groundwater. Narrow fluctuation in the standard deviation of the concentration of the heavy metals indicates low spatial variation of heavy metal concentrations in the study area. MI values display significant variation of heavy metal contamination among the wards of the Pouroshova area. Further, MI values indicate that the groundwater is slightly affected by anthropogenic sources in ward nos. 5 and 6 whereas moderately affected in the sugar mill region while the rests are pure. Similar to the MI values, the Cd index depicts worse water quality in the sugar mill region. However, the frequently used pollution index PLI interprets hardly any pollution in the groundwater of the study area although the highest negative values have found around the sugar mill area. Most importantly the HPI values confirmed that the water quality in the sugar mills region is inferior compared with groundwater of

Figure 8. Dendrogram showing the hierarchical clusters of analyzed metals.
nearby areas. The HI spatial distribution shows that infants and children are mostly in health risk to groundwater contamination at the study area while the adults are comparatively at low risk. However, a detailed study comprising more water samples from the vicinity of a group of sugar mills could draw a robust conclusion in this regard. The study findings will knock the environmental managers and policy makers to draw attention in controlling industrial pollution to save water contamination and human health.

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ORCID

Syed Md. Sazzad Hossain http://orcid.org/0000-0003-1569-1804
Md. Emdadul Haque http://orcid.org/0000-0002-7164-9638
Md. Abdul Hadi Pramanik http://orcid.org/0000-0001-6085-5949
Md. Jalal Uddin http://orcid.org/0000-0003-2640-1091

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