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Research paper

The geological setting of arsenic enrichment in groundwater of the shallow aquifers of the Tista Floodplain, Rangpur, Bangladesh

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Abstract

Arsenic is present in water samples within the studied active floodplain areas of the Tista river, Rangpur Division, Bangladesh. All the water samples contain less arsenic than the WHO prescribed limit of 10 µg/L. 93.33% groundwater samples have higher Mn content than the permissible limit of 0.01 mg/L of WHO. The heavy metal concentrations of water can be expressed as Fe>Mn>Zn>As on the basis of their mean content. The heavy metals are negatively correlated with the well depth which is indicative of the influence of the anthropogenic activities on the concentrations of heavy metals. The arsenic concentration in water samples is higher in the central part of the study area. The coarser grain size, dominance of physical weathering, elevated topography and the effective flushing of groundwater resulted low concentration of arsenic in the groundwater. The EDS study reveals that arsenic occurs as coating materials of the silicate minerals. The river waters also have arsenic content lower than WHOs permissible limit. The factor analysis reveals that the iron and arsenic is released by the chemical weathering of arsenic bearing minerals like pyrite and arsenopyrite. The Fe and Mn derived in the groundwater by the chemical weathering of iron and manganese bearing minerals such as iron rich clay, silicate minerals and iron sulfides.

Keywords: *Arsenic; Groundwater; Topography; Grain Size; Factor Analysis and Silicate Minerals.*

1. Introduction

Higher concentration of arsenic (As) in groundwater is a prime environmental problem in the aquifers of mega-deltas and low-lying flood-plains of South-east and South Asia (Fendorf et al. 2010). Studies reveal that about three million tubewells, installed at shallow depths (10 to 50m) in Bangladesh, discharge groundwater with above 50 µg/L (Ahmed et al. 2004: Zahid et al. 2008). Forty-six to fifty-seven million Bangladeshi people are exposed to drinking water containing arsenic 10 µg/L (DPHE-BGS 2001). Arsenic is present in groundwater at Siliguri-Jalpaiguri area, West Bengal, India and exactly 32% of the samples contain arsenic above the WHO prescribed limit of 10 µg/L (Bhattacharyya and Mukherjee 2008). In this area, the Tista River descending from the Himalayas and meets the alluvial plain.

The Tista and Brahmaputra River floodplains in Bangladesh have the lowest arsenic concentrations in shallow groundwaters whereas the Meghna River floodplains are reported to have high arsenic concentrations in tubewells (DPHE-BGS 2001: Ravenscroft 2001). Twenty-five percent of 524 wells beneath the Tista and Brahmaputra floodplains exceeding arsenic concentrations 50 µg/L (Ravenscroft et al. 2005). On the basis of the arsenic content in groundwater, the Tista floodplain aquifer can be categorized into two zones- the first zone which is fairly arsenic safe in the upper course in Nilphamari, Lalmonihat and Rangpur, and the next zone where 20% to 40% wells contain higher arsenic in groundwater. In particular, large part of the Tista floodplains are underlain by medium and coarse sand (Ravenscroft et al. 2005). The aim of the present research work is to determine the geochemical condition of low arsenic groundwater that was long awaited.

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2. Hydrogeology

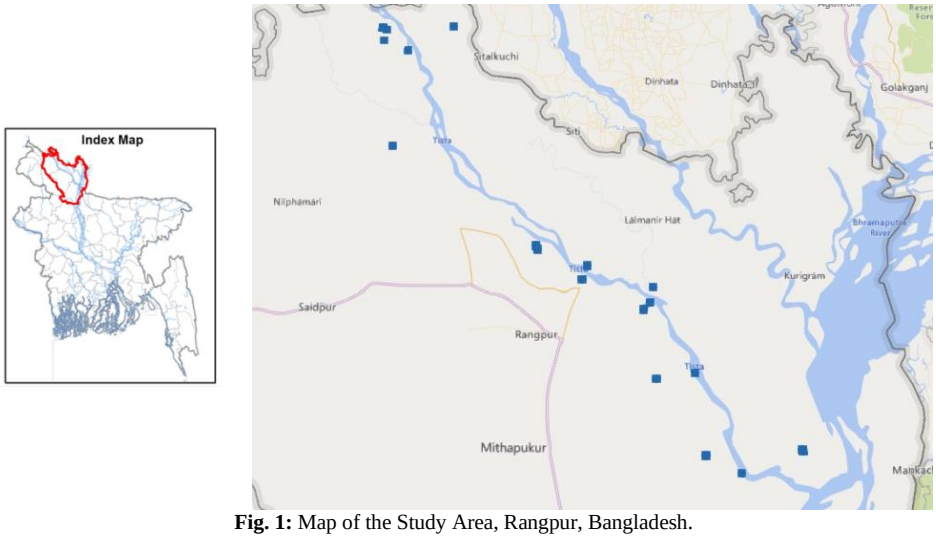


Fig. 1: Map of the Study Area, Rangpur, Bangladesh.

The Tista floodplain is characterized by big sub-region which stretches between the Old Himalayan Piedmont Plain in the west and right bank of the N-S flowing Brahmaputra in the east (Islam et al. 2016). The total catchment area of the Tista river is 12159 sq. km. According to Environmental Information System (ENVIS) centre Sikkim, total catchment area 10155 sq. km in India, and 2004 sq. km in Bangladesh. In Bangladesh the Tista river flows through Nilphamari, Lalmonirhat, Rangpur, Kurigram and Gaibandha districts of Rangpur division (Figure 1). The main tributaries of the Tista river are BuriTista and Trimohini in Bangladesh. The Tista river flows to the Brahmaputra River at Fulchary near Chilmari (Wasleker 2013). Estimates have suggested that the Tista River has a mean annual flow of approximately 60 BCM (Wasleker 2013). A significant amount of this water flows during the monsoon season i.e. between June and September. The significance of the flow and seasonal variation of this river is understood during the lean period (October to April/May) as the average flow is about 500 MCM per month. The mean annual rainfall in Northwest Bangladesh is 1,971mm (Walseker 2013). In the study area, the mean daily maximum temperature varies from 20ºC in January to 29ºC in April and June. The mean daily minimum temperatures range from 9ºC in January and February to 19ºC in July, August and October. There is a strong correlation between surface and groundwater resources in Tista floodplain (Walseker 2013). The Tista is an influent river that have a net flow from the river to the groundwater aquifer (CSIRO, WARPO, BWDB, IMW, BIDS, CEGIS, 2014).

The elevation in northwestern parts of Bangladesh (Tista Alluvial Fan) is higher than rest of the geomorphic units except the Hills of the Tertiary age (Shamsudduha et al. 2009). Most of the land shallowly flooded during monsoons.

Most of the aquifers occur at 50 cm to 130 m depth (Haque and Tasnuva, 2016). The aquifer is mainly unconfined in nature. Tista floodplain aquifers are composed of sand and gravel (Majumder et al. 2011, 2013). Rainwater is the principal source of groundwater recharge in the study area. Flood water that overflows the river and stream banks also infiltrates into the groundwater. Analysis of groundwater level and river-stage hydrographs (Shamsudduha et al. 2011) reveals that all river channels rise above groundwater levels in adjacent aquifers during the monsoon season (May-September); indirect recharge is restricted to lateral river-bank infiltration during the early monsoon time (April-June). The groundwater flows generally from north to south of the investigated area. The transmissivity values of the aquifer vary from 1000 m^2/day to 7000 m^2/day (Hussain and Abdullah 2001).

The main sources of surface water of the study area are Tista river, beels, lakes and ponds. During the summer months the flow of the Tista river is lowest and in most of the parts of the river bed dries up. The inhabitants depend mainly on groundwater for drinking and irrigation purposes. Groundwater development by tubewells requires a complete understanding of the aquifer properties, quality and quantity of the groundwater.

3. Materials and methods

From the tubewells water samples were collected in order to study physico-chemical characters of the studied water. The groundwater samples were collected from both shallow and deep tubewells. The pH, electrical conductance (EC) and temperature of the groundwater were measured and recorded in situ. Sampling was carried out using pre-cleaned polythene bottles, after pumping continuously 10-15 minutes until the temperature, electrical conductivity (EC) and pH reading had stabilized (in case of groundwater). Collected samples were preserved at 4°C and taken into the laboratory for chemical analyses. Heavy metal concentrations were determined by Atomic Absorption Spectrometer, model Shimadzu AA 7000. Thirty-five to forty sand particles were randomly selected for SEM analysis. The elemental analyses were also performed using EDS attached with scanning electron microscope.

4. Results and Discussion

The pH values of natural water vary from 6.20 to 8.50 whereas the mean value is 6.89. The pH values of the river water are higher than the pH values of groundwater samples. The median pH value of the river water samples is 8.2 that indicate the alkaline nature of the river water. The mean pH value of the groundwater 6.55 which is indicative of acidic nature of the groundwater samples of the investigated area (Saha et al. 2019).

Table 1: Statistical Summary of Heavy Metal Concentrations of Groundwater and River water, Rangpur

	Depth, m	pH	As µg/L	Mn mg/L	Zn mg/L	Fe mg/L
Mean	15.72	6.89	1.25	0.59	0.01	1.26
Median	12.20	6.60	1.25	0.54	0.00	0.75
SD	17.52	0.73	0.35	0.59	0.05	1.58
Variance	306.84	0.54	0.13	0.34	0.00	2.49
Skewness	2.50	1.24	0.05	0.77	4.26	2.00
Kurtosis	7.88	0.17	-0.97	-0.30	18.36	3.57
Max	76.25	8.50	1.87	1.91	0.21	5.74
Min	0.00	6.20	0.67	0.00	0.00	0.07
Range	76.25	2.30	1.20	1.91	0.21	5.66
Sum	298.64	130.90	23.77	11.23	0.26	23.85

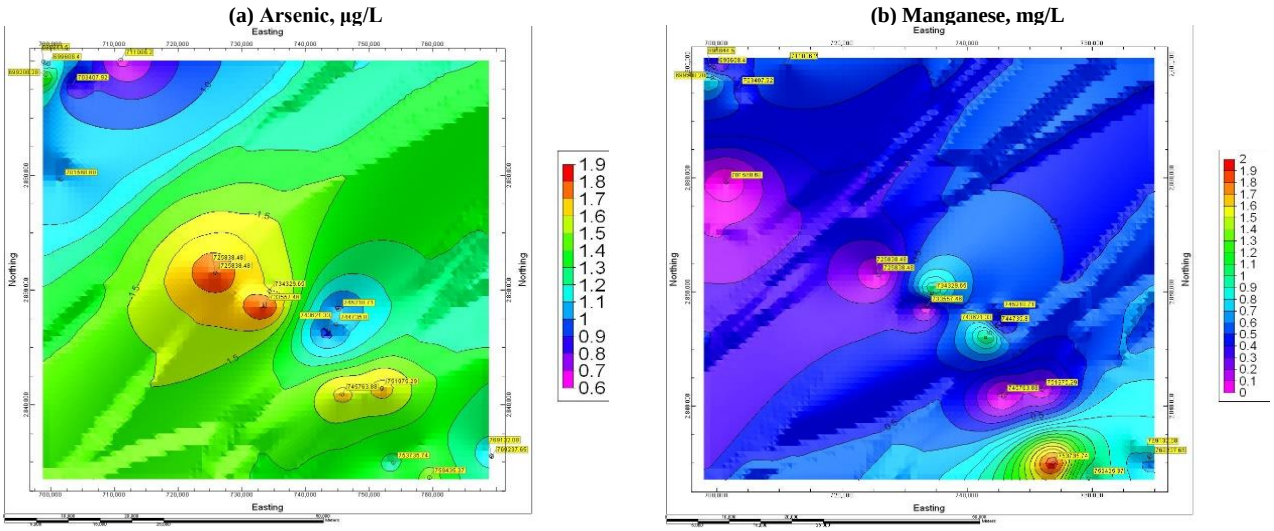


Fig. 2: Spatial Distribution of a) Arsenic and b) Manganese Concentrations of Water of the Study Area.

The average arsenic concentration of the water samples of the study area is 1.25 µg/L. The maximum and minimum values of arsenic content of the groundwater are 1.87 µg/L and 0.67 µg/L respectively. The median value for arsenic concentrations of the river water is 1.37 µg/L. The findings of the present research work reveals that the arsenic content of the water samples are below the permissible As concentration value of 10 µg/L of WHO, 2008. Figure 2a shows the spatial distribution of arsenic concentrations of natural water samples of the investigated area and it is revealed that the As concentration is highest in the central part of the study area.

The present study reveals that the Mn concentrations of groundwater samples range from 0 to 1.91 mg/L with the mean value of 0.74 mg/L. Eighty percent of the groundwater samples exceed the permissible limit of Mn, 0.10 mg/L for Bangladesh Drinking Water Standard (BD DWS) and 93.33% groundwater samples have higher Mn content than the permissible limit of 0.01 mg/L of WHO, 2011. Figure 2b shows the lateral distribution of Mn and it is clearly shown that the concentration of Mn is higher in the south eastern part of the study area. The maximum Mn content of groundwater is reported from Sundarganj Upazila of Gaibandha district. The manganese concentrations of river water samples are lower than the Mn concentrations of groundwater. The average Mn content value of river water is 0.04 mg/L.

The Zn content of all river water samples are zero. In groundwater samples, the zinc concentrations vary from 0 to 0.21 mg/L, with the mean value of 0.02 mg/L. The highest amount of zinc is reported from the groundwater of Pirgacha Upazila Rangpur district. The concentration of zinc increases to the downstream direction. The average concentration of Zn in groundwater of the study area is low.

The concentrations of iron in groundwater samples range from 0.071 mg/L to 4.790 mg/L. The mean iron concentration of groundwater is 1.16 mg/L. The iron concentrations of 1-3 mg/L can be acceptable for people drinking anaerobic well-water (WHO 2003). The present study reveals that only one groundwater sample exceeds the maximum acceptable limit of WHO, 2003. The maximum iron concentration is reported from the groundwater sample of Kaunia, Rangpur. The mean iron concentrations of river water are 1.60 mg/L, with the maximum value of 5.74 mg/L in Gangachara, Rangpur.

Table 2: Pearson Correlation Coefficient Matrix of Heavy Metals of Groundwater (n=15)

	Depth	pH	As	Mn	Zn	Fe
Depth	1.000					
pH	0.019	1.000				
As	-0.307	0.370	1.000			
Mn	-0.023	-0.151	-0.173	1.000		
Zn	-0.153	0.777**	0.335	-0.254	1.000	
Fe	-0.234	-0.129	0.207	-0.201	-0.121	1.000

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

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

The well depth shows insignificant negative correlations with As (R= -0.307), Mn (R= -0.023), Zn (R= -0.153) and Fe (R= -0.234) content of groundwater (Table 2). The negative correlations between well depth and heavy metal concentrations imply that anthropogenic activities might have influence on the concentrations of these metals in the study area.

The pH shows insignificant positive correlation with arsenic (R^2= 0.1371) and strongly significant positive correlations with zinc (R^2=0.6039) content of groundwater samples of the shallow aquifers of the Tista River Basin (Figure 3 and 4).The positive correlations between pH and arsenic are noticed from the groundwater samples of United States of America (Ayotte et al. 2003) and in Argentina (Smedley et al. 2002) and it reveals that the arsenic mobility within the groundwater is controlled by sorption/desorption processes, indicating the possibility of geologically related mineral interferences with aquifer or surface waters (Katsoyiannis and Katsoyiannis 2006).

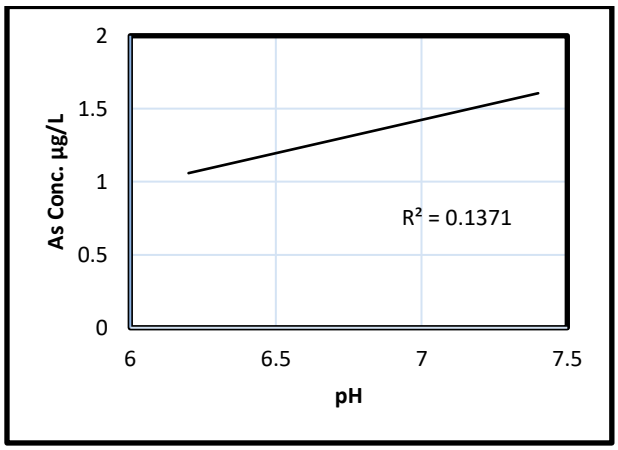


Fig. 3: Correlation of pH vs As

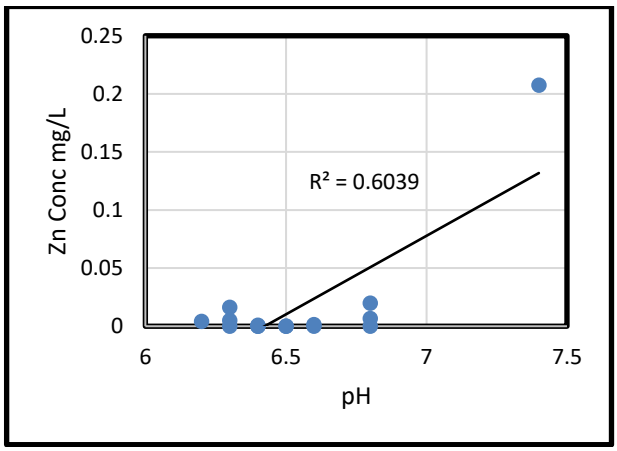


Fig. 4: Correlation of pH vs Zn

The dissolution of different arsenic bearing minerals like realgar (AsS), orpiment (As_2S3), arsenopyrite (FeAsS), enargite (Cu_3AsS_4) release arsenic into the groundwater (UN Report 2001: Moni et al. 2019). The XRD analyses of the sediments of the upper part of the aquifers reveal that they contain As-bearing mineral lavendulan [Na, Ca, Cu5 (AsO4)4 Cl. 5H2O] in trace amounts only in two locations (Saha et al. 2018, 2020).

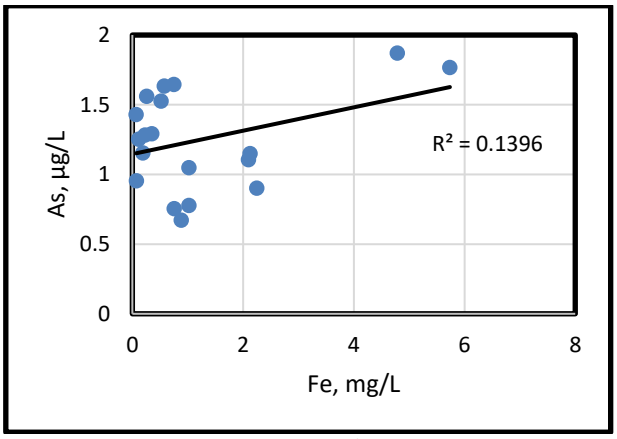


Fig. 5: Correlation of Fe vs As.

The poor positive relationship between As and Fe is observed in the investigated area (Figure 5). This poor correlation reveals that the pyrite/sulfide oxidation may release arsenic to the groundwater in small quantities (Reza et al. 2010). The arsenic concentrations in the sediments of Bengal Basin range from 0.4 to 40 mg/Kg (DPHE-BGS, 2001: Reza et al. 2010c). The geochemical study of the Tista River sediments show that the average arsenic concentration is 3.52 mg/Kg. The dominance of the coarser particles in the study area is one of the factor that result low arsenic concentrations. The illite crystallization index shows that physical weathering is the dominant in Rangpur (Saha et al, 2020). The study reveals that 93.33% groundwater samples have higher Mn content than the permissible limit of 0.01 mg/L of WHO. The insignificant negative correlation between Mn and As indicates that the source of arsenic and manganese in groundwater are different (Table 2). The EDS study shows the arsenic occurs as coating materials in platy micaceous minerals. The natural sources of iron and manganese include their derivation by the weathering of iron and manganese bearing minerals such as iron rich clay, silicate minerals and iron sulfides (Luzati et al. 2016). The shallow groundwater is free of excessive arsenic in places where the elevation is high especially in recharge areas and results remarkably higher hydraulic gradients that drive more effective groundwater flushing (Stute et al. 2007: Aziz et al. 2008: Weinman et al. 2008: Hoque et al. 2017). The present study shows that the manganese concentration of river is lower than the groundwater. The studied aquifers are in the active floodplains of the Tista River and it is an influent river, so the input of the river water into the aquifer can also lower the manganese concentration of the groundwater.

5. Factor analysis

Table 3: Total Variance Explained

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	2.207	36.789	36.789	2.207	36.789	36.789
2	1.132	18.868	55.657	1.132	18.868	55.657
3	1.008	16.792	72.449	1.008	16.792	72.449
4	.772	12.864	85.313			
5	.477	7.946	93.258			
6	.404	6.742	100.000			

Extraction Method: Principal Component Analysis.

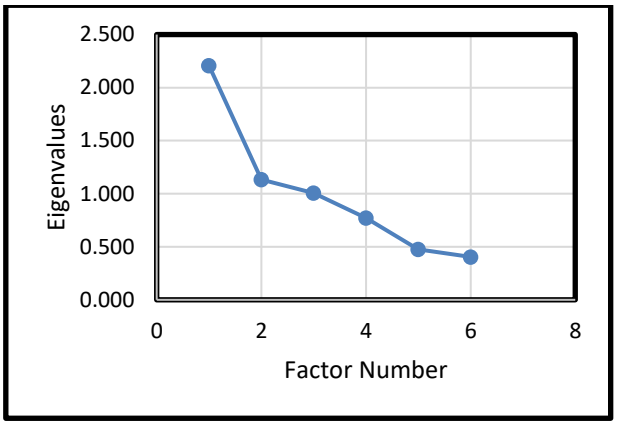


Fig. 6: Scree Plot for Heavy Minerals of the Study Area.

Table 4: Varimax Rotated Factors for First Three Components

	Component		
	Factor-1	Factor-2	Factor-3
Depth	-.665	-.127	.165
pH	.750	-.241	-.386
As	.697	.175	.503
Mn	-.653	.224	.374
Zn	.323	-.639	.628
Fe	.433	.755	.208

As describes in Table 3, the first three factors represent the ratios of 36.789%, 18.868% and 16.7922% respectively, and they comprise 72.449% of the total variability of the original data. The first factor (Factor-1) is characterized by the dominance of pH and arsenic content of the natural water and indication chemical weathering of the arsenic bearing minerals like pyrite and arsenopyrite. The second factor (Factor-2) provides data about the release of iron in the groundwater. The third factor (Factor-3) is highly affected by the contamination of zinc of the water.

6. Conclusion

Arsenic is present in water samples within the studied floodplain areas of the Tista river. All the water samples contain less arsenic than the WHO prescribed limit of 10 µg/L. The low arsenic concentration of the aquifers might have resulted as (1) the sediments have lower arsenic concentrations in comparison with arsenic affected areas, (2) the EDS study reveals that arsenic occurs as coating materials of silicate (3) prevalence of the coarser sediments in the investigated aquifers, (4) the dominance of physical weathering and (5) elevated topography and remarkably higher hydraulic gradients that result more effective groundwater flushing. The factor analysis reveals that the arsenic is released in small quantities by the chemical weathering of arsenic bearing minerals. The river waters also have arsenic content lower than WHOs permissible limit. The wells are safe considering the arsenic concentrations of groundwater whereas 93.33% of the groundwater samples have higher Mn content than the permissible limit of 0.01 mg/L of WHO. The future research works can be taken on the safe removal of manganese from the groundwater.

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