EVALUATION OF GROUND WATER CHEMISTRY IN SHALLOW AQUIFIER SYSTEM (DUGWELLS)

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ABSTRACT

Arsenic contamination is considered one of the most threatening natural calamity in Bangladesh. Different studies have been carried out to investigate the level of As contamination in shallow and deep aquifer system in Bangladesh. But no investigation has been done so far to evaluate the water chemistry or to see the level of As concentration in dugwell water. In this case, the present study is aimed to evaluate the ground water chemistry of dugwell water in South Eastern part of Bangladesh specially in Daudkandi and Sonargaon upazilla with a view to contribute to a better understanding of the complex hydrogeological and geochemical conditions.

It was found that redox potential in these two particular areas are very high indicating mildly oxidizing character of the water samples. As concentration are not significantly high which indicates that As mobilization by Fe reduction is not possible due to oxidizing environment. But some other ions like HCO_3^- and Cl^- and Na^+ and Ca^+ are very high in dugwell water.

It can be anticipated that dugwell water may be a potential option for the purpose of drinking water due to low level of arsenic concentration. However, water level in dugwells can be fluctuated with seasonal variation and this water can be contaminated easily with microbial pollutants.

KEYWORDS

dugwell, arsenic, groundwater, geochemistry

INTRODUCTION

Arsenic contamination in ground water of Bangladesh is considered one of the most threatening natural calamity currently for public health since As poisoning has several negative impacts on health and environment. Exposure to inorganic arsenic can severely affect the respiratory tract, skin, lever, cardiovascular system, hematopoietic system, nervous system, etc. (Mandal & Suzuki, 2002). There is no treatment which can cure As poisoning. It is recommended to remove exposure, treatment of skin lesions and sometimes surgery is needed if the poisoning is severe. In this case, since detected in 1993, a number of studies have been made which provide substantial data to find out the extent and distribution of the As problem. Both field test kits and laboratory analyses were conducted to get these data. Department of Public Health and Engineering (DPHE) in Bangladesh together with UNICEF field kit survey showed that 29% of the tested 50,998 wells exceeded the Bangladesh Drinking Water Standard (BDWS) which is 50 µg/l. The most systematic national survey was conducted by Bangladesh Geological Survey (BGS) and DPHE in 2001 where 3534 wells from different parts of the country were analyzed. The results indicated that nearly 27% of the shallow wells (<150 m) contain As concentrations above the BDWS, while 46% exceed the WHO provisional drinking water limit which is 10 µg/l. Approximately 3 million tubewells, placed at depths of between 10 and 50 m, yield groundwater with As concentrations exceeding the BDWS. (Matin et.al 2004).Generally groundwater from shallow alluvial aquifers(<150 m) are As contaminated and level of contamination is the maximum at a depth of 10 to 50 m. (Hasan et.al, 2008). However, only 5% of the analyzed groundwater exceeded BDWS limit for the deep tubewells (>150m). Generally, the uppermost zone of shallow aquifers (<8-10m) can be considered As safe (Hasan et.al, 2008). Therefore, most of the dugwells are constructed at this certain depth. However, there was no study done so far to evaluate the hydrogeochemical parameters of shallow aquifer system at this uppermost zone where most of the dugwells are installed. In this case, the present study has been carried out to evaluate the geochemical characteristics of dugwell water to find a correlation between the arsenic mobilization and to evaluate the hydrogeochemical and geological characteristics of the groundwater of shallow aquifer system at this zone.

Study area

The study area comprises of two upazilla in the South Eastern part of Bangladesh as shown in figure 1. They are named as Daudkandi and Sonargaon upazilla. There are approximately one million people live in these two upazillas (BBS ,2001).

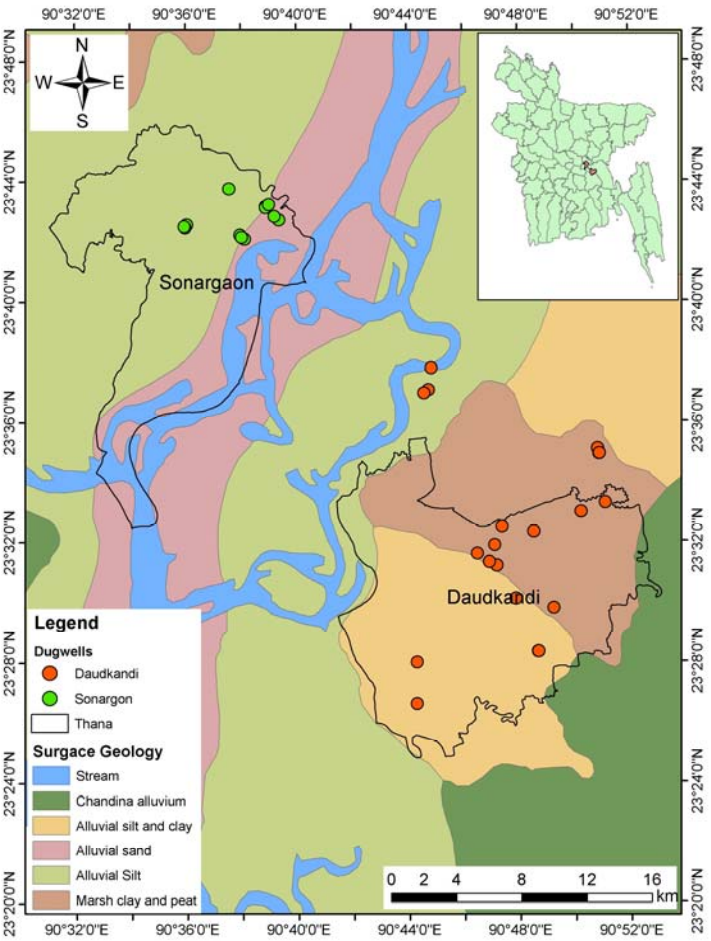

Figure 1. Geological map of Daudkandi and Sonargaon upazila in Bangladesh

Daudkandi

Daudkandi upazila is in the Comilla district lies at the east of the river Meghna in south–east Bangladesh. The area comprises of three major climatic seasons includes hot summer (March–May), followed by monsoon or rainy season (June–October) and a moderate winter season (November–February). The temperature in winter is 10 to 12 degree centigrade whereas in summer it rises up to 36 degree centigrade. The average annual rainfall in and around the study area is over 2,500 mm (according to Bangladesh Meteorology Department) and about 95% of its total rainfall is received during May to October (wet period) and rest 5% of rainfall is received during November to April (dry period). The landscape is very low lying which is situated only 1 to 4 m above sea level. The Holocene deposits that form the surface geology in and around the study area are composed of alluvial sand, silt and clay with marsh clay and peat (Fig.1). There are unconfined to leaky confined aquifer system in this area formed by unconsolidated alluvial sand. Groundwater is available within 5 m below ground surface and water level fluctuates between 2 and 4 m (Hasan et.al,2008)

Sonargaon

The climate of Sonargaon is almost same as Daudkandi. This area lies in the eastern part of the Bengal Basin west to river Meghna in South East Bangladesh. It can be said that the Bengal Basin is the most active fluviodeltaic system in the world and is tectonically divided into several uplifted and subsiding areas, including the Madhupur and Barind tracts and the Sylhet Basin. In this area Holocene deposit also form the surface geology composed of alluvial silt and alluvial sand. In this area reddish brown layers of oxidized mud and sand are exposed. Many settlements are located on these levees. (Mitamura et.al., 2008)

MATERIALS AND METHOD

Groundwater samples were collected from 25 selected dugwell sites in Sonargaon and Daudkandi upazila in Bangladesh in three different time consisting of February 2005, December 2003 and May 2004. Field parameters (pH, redox ,conductivity and dissolved oxygen)were measured in a flow through cell preventing oxygenation of the samples. The pH was measured using a Hanna onsite pH meter combination of electrode and the redox potential was measured using a combined platinum electrode(MC408Pt) equipped with a calomel reference electrode. Field measured redox potential was corrected for the standard hydrogen electrode(SHE). The electrical conductivity was measured using a HANNA using HI7833 conductivity meter with an operating range between 0 to 200 mS/cm. Dissolved oxygen was measured using a Luton (DO 5510) meter. After the onsite measurement the samples were stored in 30 ml polyethylene bottles following the sampling protocol described by Bhattacharya et al. (2002) and Hasan et al. (2007). All the samples were filtered in the field using Sartorius 0.45 µm single use syringe filters. Samples for cation and trace element analyses were acidified with suprapure HNO3 (14 M) to bring the pH < 2 and samples for anion analyses were preserved without acidification. The bottles were capped tightly and shipped by air to Stockholm for chemical analyses after coming back from the field. Samples were stored at controlled temperature for 1–2 months prior to analysis. However, the storage conditions were beyond control during the air shifting time. Ammonium and dissolved organic carbon (DOC) were determined at the water chemistry laboratory of the Royal Institute of Technology. Stockholm. Sweden. Ammonium was analyzed using spectrophotometer with Tecator Aquatec 5400 at 540 nm wavelength. Alkalinity was determined by titration according to the standard method SS-EN ISO 9963–2 (SIS. 1996) using Radiometer

Copenhagen PHM 82. Dissolved organic carbon (DOC) in the water samples were determined on a Shimadzu 5000 TOC instrument. Cations ($Ca^{2+.}$ $Mg^{2+.}$ Na^+ and K^+) and trace elements including total arsenic were analyzed by inductively coupled plasma (ICP) emission spectrometry (Varian Vista-PRO Simultaneous ICP-OES equipped with SPS-5 auto sampler) at Stockholm University, Sweden.

RESULTS AND DISCUSSIONS

On-site field parameters

In general, measured electrical conductivity of all water samples reveals moderately high values. However, the highest value of 2430 μS/cm observed in samples collected from Daudkandi and 2700us/cm in Sonargaon..pH values of groundwater samples both in Sonargaon and Daudkandi upazilla is slightly acidic and in some places it is nearly neutral. pH(6.5–7.04) with low dissolved oxygen (1.74 to 0.99 mg/l). The Eh values (+185 to 366 mV) shows mildly oxidizing character of the aquifers in Daudkandi and Eh values (+259 to 391 mV) in Sonargaon area also follow the same mild oxidizing character. Average temperature found in Daudkandi is 26^0C and for Sonargaon 25.4^0C.Groundwater in both the upazilla is generally Ca– Mg-HCO_3 or Ca–Na–HCO_3 type. Regarding different water type here it can be anticipated that Ca dominating water type is very significant compared to Na dominating water types.

Major ion composition

Table 1 and Table 2 show the major cation and anaion composition in Daudkandi and Sonargaon upazilla. It can be anticipated that Dissolved ions in both areas are dominated by major cations (Ca+ and K+) and major anion (HCO_3^- and Cl-). The average value for HCO_3^- is 330mg/l for Daudkandi and 291 mg/l for Sonargaon upazilla. The average value for Cl^- is very high and almost similar for both upazila which is 150mg/l for Daudkandi and 147.77 mg/l for Sonargaon. Distribution of the major cations such as Ca (40–170 mg/l). Mg (14–41 mg/1), Na (7–150 mg/1) and K (2-125mg/l) is significantly high in Daudkandi as well as in Sonargaon. In Sonargaon the cation concentration varies considering Ca (32-194 mg/l), Mg(9-63mg/l) and Na (12-177 mg/l) and K (3–127 mg/1).This concentration show a significant variations with depth as well as region. The average concentrations of SO_4^{2-} is 72 mg/l considering the maximum value of 167 mg /l in Daudkandi and the average value is 62.15mg/l with a maximum value of 153 mg/l in Sonargaon.. Median values for Ca is high for both upazila (74.3 mg/l in Daudkandi and 72 mg/l in Sonargaon) but the concentration varies for Mg which is 39.4 mg/l for Daudkandi whereas 20mg/l for Sonargaon only. The median values for HCO_3 352.7 mg/l for Daudkandi and 300 mg/l for Sonargaon upazilla. It was found very high concentration of HCO3- in dugwell water which can be attributed to: (i) high OM releasing HCO_3.Another possibility of high HCO3 concentration may be due to carbonate dissolution.

Table 1. Statistical analysis of major ion composition in Daudkandi (n=14)

Parameter	Unit	Min	Max	Median	Average	Q25	Q50	Q75	Q90
Ca	mg/l	41	170	74.3	85.78	57	69	95.5	135
Mg	mg/l	17	91	39.4	43.64	33	38	54	61
Na	mg/l	14	102	52.1	56.14	37.5	52	74.5	78
K	mg/l	2	125	8.7	39	5	7	63	120
HCO_3	mg/l	175	485	352.7	330.93	237	332	388	447
Cl	mg/l	35	436	122	150	62	122	197.5	238
NO_3	mg/l	0	75	19.4	23.714	0.5	19	36	44
SO_4	mg/l	1	167	92.9	72	15.5	92	99.5	112

Table 2. Statistical analysis of major elements in Sonargaon(n=13)

Parameter	Unit	Min	Max	Median	Average	Q25	Q50	Q75	Q90
Ca	mg/l	32	194	72	86.53	41	67	122.5	142
Mg	mg/l	9	63	20	29.38	15.25	19.5	35.5	59
Na	mg/l	12	177	51	61.85	31	50	74.5	90
K	mg/l	3	127	32	34.39	10.75	24.5	40.25	56
HCO_3	mg/l	145	367	300	291.38	258	287	348.5	357
Cl	mg/l	23	493	97	147.77	69.25	89.5	162.25	279
NO_3	mg/l	0	77	10	14.77	4	7	14.25	27
SO_4	mg/l	4	153	46	62.15	20.25	42	83.5	139

Redox sensitive parameters and other trace elements

Average concentration of total arsenic [As(tot)] 8.74 µg/L in Daudkandi and 9.0µg/L in Sonargaon.(Table 3 and Table 4). There were few extremely high values. The maximum concentration of As was 59.µg/L in Laxmibardi in Daudkandi and 64.68 µg/l in Ladurchor in Sonargaon. The median concentration of As(tot) was (8.74 µg/l) in Daudkandi whereas the median concentration in Sonargaon was 2.80 also which is same as the minimum concentration in this upazila. Arsenic concentration in some of the samples in the study area was below detection limit and in some cases the concentration was quite near to detection limit and some places that is above detection limit. However with some few exception for example in Ladurchor and Laximbardi the As concentration is quite high. Otherwise this study is sowing that arsenic concentration in this area for dugwells is insignificant compare to deep aquifer system. Manganese concentration (average 504 µg/l. median 1171 µg/l) is very high in Daudkandi as well as in Sonargaon which is (median 276 µg/l and average 785 µg/l. The concentrations of Fe (medean 459 µg/l. average 140 µg/l) in Daudkandi were similar to those of Mn in The groundwater. Significant variations in the concentrations of total iron (Fetot) and Mn^{2+} has been observed within the water samples in Daudkandi and Sonagaron upazilla. The average values of iron in groundwater samples are 782 and 221 µg/l. Higher values of Mn2+ suggest an on-going Mn-reduction in the samples according to this equation: $CH_2O + 2\ MnO_2 + 4\ H^+ \rightarrow CO_2 + Mn^{2+} + 3\ H_2O$ (Bakr. M., 1977).). Here manganese is redox-buffering the system (i.e. may be $Mn^{2+}MnO_2$ pair preventing significant change in Eh) and thus restricting the groundwater system to reach the stage of iron reduction. The concentrations of the analysed trace elements and their statistical summary is presented in table 4 and table 5.

Table 3. Concentration of different trace elements in Daudkandi upazilla (n=14)

Parameter	Unit	Min	Max	Median	Average	Q25	Q50	Q75	Q90
As	µg/l	2	59	8.74	8.29	2	2	2	19
Fe	µg/l	27	5680	88.53	782.71	31	49	284.5	2398
Mn	µg/l	0	6933	504.18	1171	124.5	492	785	2928
DOC	mg/l	0.93	5.07	2.54	2.67	1.61	2.54	3.359	4.366
PO4	µg/l	0	7817	200	1124.5	110.5	198	888	2098
Cu	µg/l	0	40	1.8	5.2	0	1	2	13
Al	µg/l	8	21	12.38	12.28	9	11	14.5	16
Li	µg/l	0	15	5.73	4.93	1	3	7.5	10
Si	µg/l	23	47	39.41	37.5	31.5	39	41	47

Table 4. Concentration of different trace elements in Sonargaon upazilla(n=13)

Parameter	Unit	Min	Max	Median	Average	Q25	Q50	Q75	Q90
As	µg/l	2	64	2.8	9	2	2	5.75	13
Fe	µg/l	21	1200	82.98	221.92	40.75	81	140	584
Mn	µg/l	18	2554	276.57	785.84	112	269	1437.5	1785
DOC	mg/l	0.81	5.11	1.8	2.22	1.48	1.8	2.91	3.21
PO_4	µg/l	0	6717	0	1603.77	29.5	276.5	2098.75	5234
Cu	µg/l	0	1	0.39	0.31	0	0	0.75	1
Al	µg/l	7	15	9.16	9.31	8	8.5	9.75	11
Li	µg/l	1	18	6.84	7.69	4	6	10	13
Si	µg/l	26	54	35.83	38.53	31	34.5	42	52

Phosphate (PO_4^{3-}) concentrations also vary between the two upazila (Table 4 and Table 5). The average values PO_4^{3-} in water derived from Daudkandi water sample is 1124 and 1603 µg/l Higher concentrations of PO_4^{3-} can be attributed to the precipitation of different mineral phases (e.g. vivianite. $Fe_3(PO_4)_2 .8H_2O$) during the dissolution of Fe-oxy-hydroxides. Very low nitrate (NO^{-3}) concentrations in all the groundwater samples (maximum 2 mg/L) may be due to nitrate reduction through ammonification reaction and denitrification processes which can be showed as:

$$CH_2O + 4/5\ NO^{3-} + 4/5\ H^+ \rightarrow 2/5\ N_2 + CO_2 + 7/5\ H_2O.$$

While considering the concentration of DOC, it was found that groundwater from Daudkandi contain high DOC concentrations (median 2.54 mg/L for Daudkandi and 1.8 mg/L for Sonargaon). Maximum concentration of DOC is of 5.07 mg/l for Daudkandi and 5.11 mg/l for Sonargaon. Although DOC characteristics in Bangladesh ground waters are not investigated in detail. However, high DOC concentrations may be due to the presence of high organic content in these Holocene sediments (Nickson et al. 2000; Bhattacharya et al.. 2002; Ahmed et al.. 2004; McArthur et al. 2004). It is still not clear whether organic matter (OM) has been formed due to the decomposition of peat layers or from seasonal water-level fluctuations of agricultural and other organic wastes from near-surface environments.

Regarding the concentration of trace elements it can be said that high concentrations of dissolved Si in the range from 11.6 to 43.8 µg/l (mean of 28.1 µg/l) has been found in Daudkandi..

Among the trace elements. the concentrations of. Li (43.5–757 µg/l; mean 189 µg/l) were also high. The concentration of dissolved Al varied between 0.0 and 7.82µg/l with an average value of 1.1 µg/l in Daudkandi. However. The average concentration of Al decreases significantly (median 12.38 µg/l in Daudkandi and 9 µg/l in Sonargaon)

Correlation between various chemical parameters

A very strong correlation between DOC and HCO_3 have been found. The saturation index for carbonate minerals (e.g. calcite. aragonite. dolomite) is close to zero. However. carbonate dissolution cannot be the only source of HCO_3^- in groundwater. Iron indicates moderately strong positive correlation with HCO_3^- and $PO_4{}^{3-}$ while it exhibits a relatively low correlation with Astot .This is commonly expected in reducing groundwaters with elevated HCO_3^- and PO_4^{3-} levels where Fe^{2+} may precipitate as siderite ($FeCO_3$) and or otherwise vivianite ($Fe_3(PO_4)_2.8H_2O$. A weak negative correlation between As_{tot} with HCO_3^-indicating a highly oxidizing condition in dugwell water samples. Since no strong correlation have been found with different As and other redox sensitive parameters therefore. it could be assumed that As mobilization is not so significant but still needed to be studied further. We know the major mechanism behind As release can be attributed to reductive dissolution of Fe-oxyhydroxides. In this dugwell water samples the oxidation potential is very high which means the water is more attributed to oxidizing environment since the depth of dugwells is only 6 to 8 meter below ground level. Therefore, reductive dissolution of Fe is not possible and hence the As concentration is not significant in this particular area.

Figure 2 shows the different correlation between As with other redox sensitive parameters in these two upazillas.

There is no significant correlation found between Mn and DOC .The weak correlation r2 =0.3 for Dadukandi and 0.045 for Sonargaon implies that there is no significant influence of DOC with Mn concentration.

Bicarbonate (HCO_3^-) concentrations in water samples derived from ground water show strong positive correlation (r^2=0.76) with DOC for Daudkandi and r^2=0.14 for Sonargaon. (Fig. 2). Thus. the process of generation of high DOC due to biodegradation of OM and then HCO_3^- in response to other redox reactions e.g. iron reduction is mainly confined to the recent Holocene black sediments. There is no significant correlation derived from correlation between Astot and HCO_3^- in water samples (r2=0.18 in Daudkandi and r2= 0.19 in Sonargaon) .There is moderately strong correlation has been found between Fe and DOC and Mn and DOC.

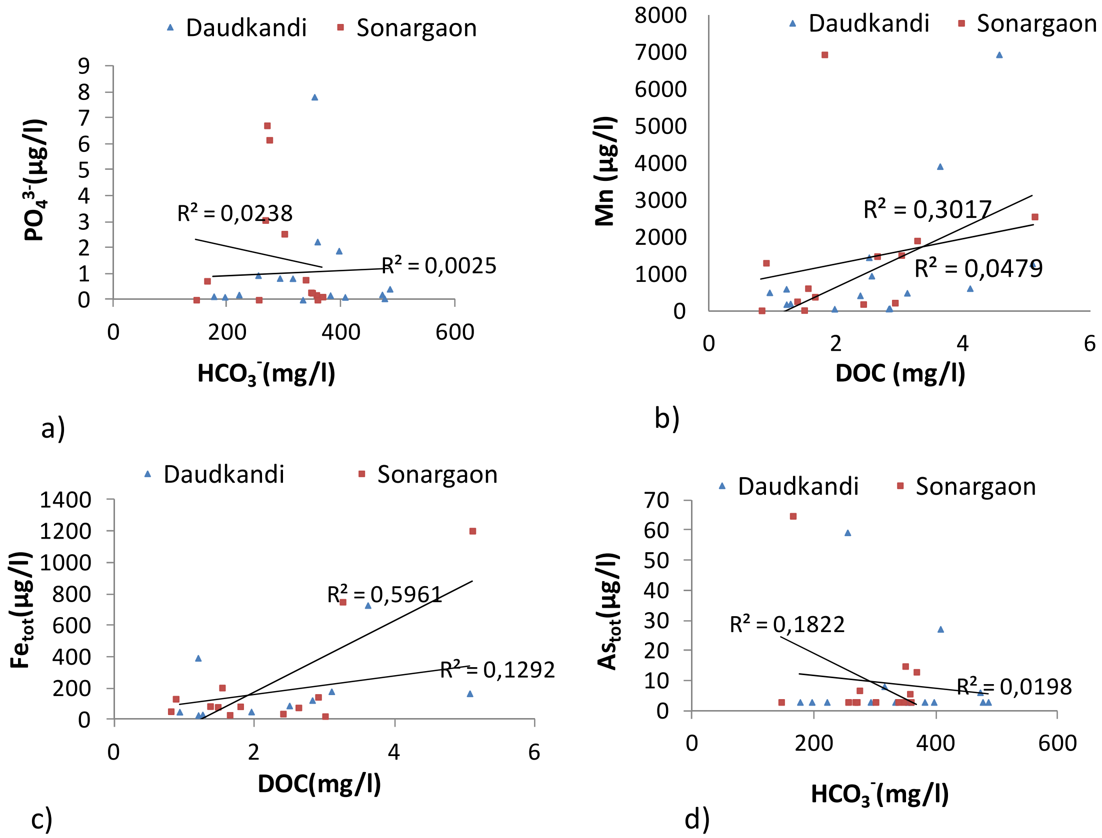

Figure 2. Correlation between different redox sensitive parameters in Daudkandi and Sonargaon upazilla a) PO_4^{3-}vs HCO_3 b)Mn vs DOC. C)HCO3 vs DOC. and d)As tot vs HCO_3^-

Speciation modelling

Figure 3 shows the Saturation Index (SI) of average values of different water sample in Daudkandi and Sonargaon. It is shown that with respect to carbonate phases; e.g. calcite ($CaCO_3$). dolomite [$CaMg(CO_3)_2$]. and aragonite ($CaCO_3$) suggests low to moderate HCO_3 concentrations which is not only due to carbonate dissolution (Equation II). but also due to oxidation of OM (Ahmed et al.. 2004). Fe^{2+} was found as the major aqueous species of Fe and thus groundwater is supersaturated with respect to Fe (III) phases like ferrihydrite [$Fe(OH)_3$]. goethite (FeOOH). hematite (Fe_2O_3) (Fig.3) and magnetite (Fe_3O_4) (); in other words. The concentration of Mn^{2+} is very high in this particular region which can be attributed to precipitation of Mn (II) minerals like rhodochrocite; but groundwater is slightly under-saturated to near-equilibrium with respect to rhodochrocite. The principal species of Fe is Fe^{2+}. of samples is supersaturated with respect to siderite indicating precipitation of Fe (II) phases is favorable in this area.

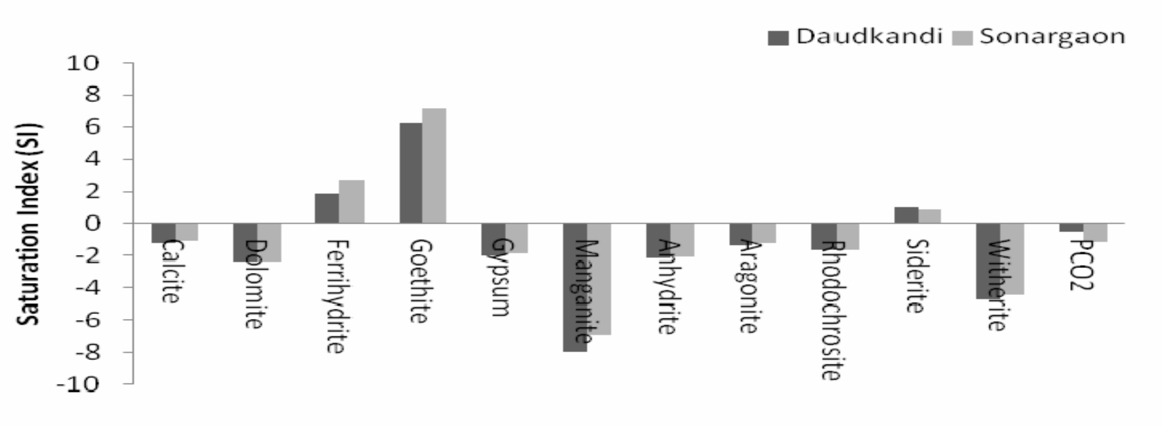

Figure 3. Saturation indices for various minerals present in Daudkandi and Sonargaon

Both Sonargaon and Daudkandi water sample shows very high supersaturated concentration of hematite which indicates the oxidizing environment. Some samples are slightly supersaturated with respective to siderite ($FeCO_3$) suggesting that precipitation of Fe(II) phases is favorable. All of samples are slightly under-saturated with respect to rhodochrocite ($MnCO_3$) and highly under-saturated with respect to Mn-oxides and hydroxides; manganite (MnOOH). (Fig.3). Precipitation of rhodocrocite might have played a significant role in removal of Mn from the solution (Mukherjee et al.. 2008). However, groundwater is highly under saturated with manganite. No significant variation can be observed in P_{CO2} values within the water samples in Daudkandi and Sonargaon.

CONCLUSIONS

Shallow groundwater system specially the geochemical characteristics of dugwell water is relatively poorly understood in Bangladesh. Though shallow ground system can be treated as potential option for arsenic mitigation since in our study it is obvious that arsenic concentration in most samples were not so significant. Some specific evaluation of hydrochemcial characteristics have been performed and following salient features have been observed.

We know the major mechanism behind As release can be attributed to reductive dissolution of Fe-oxyhydroxides. In this dugwells, the oxidation potential is very high which means the water is more attributed to oxidizing environment since the depth of dugwells is only 6 to 8 meter below ground level. Therefore, reductive dissolution of Fe is not possible and hence the As concentration is not significant in this particular area

There was found very concentration of HCO_3^- which can be attributed to: (i) high OM content in the ground water which may derive from the decomposition of peat layer.

Regarding use of dugwell as a source for As safe drinking water but it can be said that this water may be contaminated easily by microbial pollutants even though it may not be attributed to high level of arsenic contamination. Shallow groundwater system is more susceptible to water level fluctuation due to seasonal variation. For instance in summer season the water level drops down so that the well gets dry and therefore there is no access to ground water. In this case microbial contamination may not be possible. Therefore, more research should be carried out and few steps should be taken in achieving arsenic- and bacteria-free water by selecting a site suitable for a dugwell and by following strict adherence to sanitary standards.

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