Geochemical assessment of groundwater composition and evaluating its suitability of Saidpur Upazilla in Bangladesh

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Abstract: The chemical analysis of 44 groundwater samples in the northern Bangladesh has been evaluated to determine the hydrogeochemical processes and major ions, heavy and rare metal concentration for its suitability for agricultural and domestic purposes. The quality analysis is performed through the estimation of pH, EC, cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, Zn^{2+}, Cu^{2+}, Mn^{2+}, Fe^{3+} and As^{3+}), anions (CO_3^{2-}, HCO_3^-, NO_3^-, SO_4^{2-}, PO_4^{3-} and Cl^-) and TDS (total dissolved solids). We also computed several variables such as SAR (sodium adsorption ratio), SSP (soluble sodium percentage) RSC (residual sodium carbonate), potential salinity, permeability index, Kelly's ratio, Gibbs ratio and hardness to evaluate the suitability of groundwater supply for specific uses. From the geochemical results, it has been found that both the cations and anions varied in the groundwater. Among the chemical budget of ions, magnesium and chloride were found to be the most predominant ions. The intense agricultural activities may be an important factor for the higher concentration of nitrates in these aquifers. Based on the total hardness, most groundwaters are moderately hard. According to EC and SAR the most dominant class is C1-S1. The major ion concentrations are below the acceptable level for drinking water. The salinity hazard is low thus, there is less chances to increase of toxic salt concentrations. Gibbs diagram indicates that all the samples fall in the precipitation dominance field. Regarding cation and anion constituents, groundwater is suitable for irrigation and drinking purposes except of few wells.

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Introduction

Groundwater is an important resource of freshwater for agricultural, industrial and drinking purposes in Bangladesh. Water of adequate quantity and quality is required to meet growing household, industrial, and agricultural needs (Azaza et al., 2010). Increasing populations, food insecurity, growing economies and poor water management are putting unprecedented pressure on the world's freshwater resources. As population grows, demands on groundwater and surface water resources are increasing worldwide in arid and semi-arid regions (Meigh et al., 1999). With the increasing competition for freshwater, there is a growing reliance on the abstraction of groundwater for irrigated agriculture. Groundwater irrigation demand has been growing steadily over the past decades, for many reasons including the unreliability of the traditional large canal schemes, and the increasing need of farmers to manage their own irrigation applications. In addition, unpredictability in climate has forced some farmers, particularly in northern Bangladesh, to exploit groundwater, in order to combat drought. The increasing overexploitation of important aquifers around the world, as well as groundwater contamination must be of concern to water resource planners and managers. Groundwater is becoming the most essential input for increasing crop production as well as for the sustainable agricultural development. Availability of groundwater for irrigation has contributed to manifold increases in crop productivity in Bangladesh. The ratio of groundwater to surface water use is much higher in northwestern districts of Bangladesh compared to other parts of the country.

All the rivers and cannels of the area dry up during the dry season and make the people completely dependent on groundwater (Shahid and Behrawan, 2008).

Natural chemical composition of groundwater results from two main processes: the first is the atmospheric salts that coming from marine aerosols, dust and concentration by evaporation of dissolved salts in precipitation, and the second are the interaction of groundwater with the formation minerals (Jordana and Batista, 2004). In aquifers expose to human activity, the quality of water can be directly affected by the infiltration of pollutants in the recharge area of aquifers (Daghrah, 2009; Daghrah and Al-Sa'ed, 2009). In addition to natural sources, groundwater quality could be affected by urbanization, agricultural waste, land cover, intensive applications of fertilizers, pesticides, utilization of wastewater for irrigation, leakage from wastewater lagoons, landfill disposal sites, septic tanks and industrial discharge (Scanlon et al., 2005; El-Naqa et al., 2007; El-Saeid et al., 2011). The groundwater in different country was contaminated probably due to lack of proper waste management (Nkolika and Onianwa, 2011). According to the previous investigation, groundwater at some locations if used for irrigation may cause either soil salinity or sodicity problems due to high salt concentration and high sodium contents relative to other elements (Al-Naeem, 2011). However, specific water may be suitable for irrigation but may not be suitable for drinking and industrial uses due to presence of some other ions at toxic level (Tanninen et al., 2005).

Bangladesh is located in a sub-tropical area with an average annual precipitation less than one third of that of the world. Furthermore, spatial and temporal distribution of the regional precipitation is not integrated and also these resources are at greater risk to contamination. Yet few studies related to groundwater quality and irrigation practices are available for this region. Studies in Bangladesh have (Hakim et al., 2009; Islam et al., 2010; Jahidul et al., 2010) showed that HCO_3^-, Na^+, Ca^{2+}, Mg^{2+} and Cl^- were the dominant ions in groundwater collected from different regions. Intense agricultural have placed a high demand on groundwater resources in northern region of Bangladesh. In addition to elevated concentrations of nitrate, chloride, phosphorus, and pesticide residues in groundwater, there may be significant temporal and spatial variability in groundwater chemistry beneath agricultural land (Delin and Landon, 2002). Understanding the quality of groundwater is as important as its quantity because it is the main factor determining its suitability for drinking, domestic, agricultural and industrial purposes. The evaluation and management of groundwater resources require an understanding of hydrogeological and hydrochemical properties of the aquifer. Therefore, the hydro geochemical investigation was carried out to identify groundwater geochemistry and its suitability for irrigation and drinking purposes.

Materials and methods

Geographic and geologic description of the area

The study area is located (25° 44′ to 25° 52′N, 88° 52′ to 89.00′ E) in Saidpur Upazilla in the northern part of Bangladesh, and has an area of approximately 121 km^2. Bangladesh has a tropical monsoon climate characterized by heavy seasonal rainfall, high temperatures and high humidity. The average rainfall during monsoon ranges from 1194 mm to 3454 mm. Another feature characterizing the precipitation in the study site is its irregular yearly distribution. The area has a sub-tropical climate, with mean maximum summer temperatures (July) about 40°C and minimum winter temperatures (January) of 10°C. The area has complicated land use characteristics, mainly consisting of agricultural and residential areas.

The lithology of most northern region of Bangladesh, especially this study area, consists predominantly of medium to coarse grained, poorly sorted sands and gravels with thin surface clays. A previous UNDP study classified the groundwater aquifers of Bangladesh into three zones, namely the upper aquifer, main aquifer and deep aquifer (UNDP, 1982). We collected the water samples from the main aquifer. This is the main water bearing zone and occurs at depths ranging from less than 5 m in the northern region of Bangladesh. This aquifer is either semi-confined or leaky, and consists of stratified interconnected, unconfined water bearing zones. The aquifer is comprised of medium and coarse grained sediments, in places inter-bedded with gravel (UNDP, 1982). These sediments occur to depths of about 140 m below ground surface.

Sampling and analytical methods

A total of 44 groundwater samples (16 shallow tubewells, 12 hand tubewells and 16 deep tubewells) were collected from existing wells in the study areas in March and April 2012 (Fig. 1). Well water samples were collected during the time period when groundwater levels are generally lower relative to other seasons of the year (Shamsudduha et al., 2009). Samples were collected in two liter plastic bottles that had been cleaned with hydrochloric acid (1:1) and then rinsed with tap water followed by rinsing with distilled water. Before collecting each sample, bottles were rinsed 3 to 4 times with sample. Samples were analyzed in Department of Agricultural Chemistry, Hajee Mohammad Danesh Science and Technology University, Dinajpur, Bangladesh. All reagents are of analytical grade, purchased from Aldrich chemical company, England. For SO_4^{2-} and NO_3^-, samples were refrigerated and analyzed within 24 h. For heavy metals analysis, samples were filtered immediately using 0.45 µm filter paper. The filtrates were acidified to pH = 2 with nitric acid in order to keep the metals in solution.

In order to assess the suitability classes for irrigation, domestic and industrial uses, we measured pH, EC, TDS, Ca^{2+}, Mg^{2+}, Na^+, K^+, Zn^{2+}, Cu^{2+}, Mn^{2+}, Fe^{3+}, PO_4^{3-}, As^{3+}, CO_3^{2-}, HCO_3^-, SO_4^{2-}, NO_3^- and Cl^-. The pH and conductivity were measured using pH meter (Orion Research, Model SA 520, USA) and conductivity meter (JENWAY, Model 4010, UK), respectively. TDS was measured by drying and weighing method. Zn^{2+}, Cu^{2+}, Mn^{2+} and Fe^{3+} were analyzed by atomic absorption spectrophotometry (APHA, 1998) in the Soil Resources Development Institute, Dinajpur, Bangladesh. Arsenic was determined by atomic absorption spectrophotometer equipped with hydride generator (APHA, 1998). A Perkin–Elmer Analyst 100 atomic absorption spectrophotometer equipped with a FIAS-100-flow injection hydride generation system was used for arsenic measurements. All samples for arsenic determination were pre-reduced with concentrated HCl (1 ml), 5 % KI and ascorbic acid mixture prior to hydride generation. Ca^{2+} and Mg^{2+} were analyzed by complexometric titration. K^+ and Na^+ were estimated by flame emission spectrophotometry. SO_4^{2-} was determined turbidimetrically. CO_3^{2-} and HCO_3^- were analyzed titrimetrically. Chloride was estimated by argentometric titration (APHA, 1998) and PO_4^{3-} and NO_3^- were determined colorimetrically (APHA, 1998). The precision of measurements was checked taking three replicates from the sample. The accuracy of the analysis for major ions was cross checked from the electrical balance, since the sum of positive and negative charges should be equal.

Whether groundwater is suitable for a particular purpose depends on the criteria or standards of acceptable quality for that specific use. The following formulae related to the irrigation water classes rating were used to classify water samples using the chemical data.

a) Sodium Adsorption Ratio (SAR)

$$SAR = \frac{Na+}{\sqrt{\frac{Ca^{2+} + Mg^{2+}}{2}}}$$

b) Soluble Sodium Percentage (SSP)

$$SSP = \frac{\text{Soluble Na concentration (meq/L)}}{\text{Total cation concentration (meq/L)}} \times 100$$

c) Residual Sodium Carbonate (RSC)

$$RSC = (CO_3^{2-} + HCO_3^-) - (Ca^{2+} + Mg^{2+})$$

d) Hardness or Total Hardness (H_T)

$H_T = 2.5 \times Ca^{2+} + 4.1 \times Mg^{2+}$ (Freeze and Cherry, 1979)

e) Potential salinity = $Cl^- + (SO_4^{2-}/2)$

$$\text{Permeability index (P.I)} = \frac{Na^+ + \sqrt{HCO_3^-}}{Ca^{2+} + Mg^{2+} + Na^+}$$

f)

Kelly's ratio=$Na^+/(Ca^{2+} + Mg^{2+})$

g) Gibbs Ratio I (for anion) = $Cl^-/(Cl^-+HCO_3^-)$

Gibbs Ratio II (for cation) = $Na^++K^+/(Na^++K^++Ca^{2+})$

where concentrations of ionic constituents for calculating all parameters are in meq/L except hardness (mg/L).

Results and discussion

Chemical composition of water samples

Details of the sampling sites are presented in Fig. 1 and Table 1. Some summary results from our survey of groundwater are shown in Tables 2 and 3. The pH of the samples is acidic in nature (pH range from 3.8 to 6.1; Table 2). Electrical conductivity is significantly affected by the temperature, so all results were normalized to a standard temperature of 25°C. A high concentration of salts in irrigation water renders the soil saline. This also affects the salt intake capacity of the plants through the roots. Specific conductance of water samples ranges from 80 to 790 µS/cm in the study area. The EC values were distinctly dissimilar among the samples. The TDS ranged from 54 to 529 mg/L, respectively. Groundwater contain higher TDS due to appreciable amount of dissolved bicarbonate, chloride and sulfate compounds containing Ca^{2+}, Mg^{2+}, Na^+ and K^+ (Karanth, 1994). NO_3^- and SO_4^{2-} concentrations were 0.10 to 1.65 mg/L and 0.003 to 1.08 meq/L, respectively. Concentrations of Na^+ and K^+ ranged from 0.098 to 1.45 meq/L and 0.009 to 0.33 meq/L, respectively. Potassium concentrations were generally lower than Na concentrations. Ca^{2+} and Mg^{2+} were major cations in groundwater and ranged from 0.08 to 2.73 meq/L and 0.81 to 4.37 meq/L, respectively. An appreciable amount of HCO_3^- was present in all water samples, though CO_3^{2-} was negligible in most cases. The range for HCO_3^- were 0.047 to 2.25 meq/L while Cl^- concentrations ranged from 0.25 to 3.00 meq/L. Fe^{3+} and Mn^{2+} concentrations varied from 0.038 to 2.62 mg/L and 0.003 to 2.25 mg/L, respectively (Table 3). Arsenic concentrations ranged from 0.008 to 0.032 mg/L. The chemical compositions of both the cations and anions were in agreement with some other reports (Jahidul et al., 2010; Islam et al., 2010; Hakim et al., 2009) The computed variable, hardness, varied from 52 to 428 mg/L. Kelly's ratio varied from 0.03 to 0.20 (Table 4). The spatial variation of chemical parameters in the groundwater reflects the natural and human activity variation. The relative content of a cation or an anion is defined as the percentage of the relative amount of that ion to the total cations or anions, respectively.

The concentrations of Ca^{2+}, Mg^{2+}, Na^{+} and K^{+} represent on average 31.29, 53.93, 13.23 and 1.55% of all the cations, respectively. Among the anions, the concentrations of Cl^{-}, HCO_3^{-}, SO_4^{2-}, and NO_3^{-} represent on average 51.25, 18.53, 8.02 and 22.20%, respectively. Thus, the order of cation and anion abundance is $Mg^{2+} > Ca^{2+} > Na^{+} > K^{+}$ and $Cl^{-} > HCO_3^{-} > NO_3^{-} > SO_4^{2-}$, respectively.

Factors controlling the composition of groundwater

Relationships between major cations and anions have been employed to deduce the probable sources of these ions in groundwater. The chemical composition of groundwater evolves during regional flow, and this evolution can be generalized by considering the water types that are typically found in different zones of groundwater flow systems (Ingerbritsen et al. 2006). In the study area, the evolution of the chemical composition in the shallow groundwater is affected by climate, hydrology, geomorphology, lithology, and other comprehensive factors. The dominant controlling factors of water chemistry vary from region to region. The Na-Cl relationship has often been used to identify the mechanisms for acquiring salinity and saline intrusions (Jalali, 2007). Most groundwater samples in this study had Na^{+}:Cl^{-} ratio lower than unity, while a few had Na^{+}:Cl^{-} ratio equal to one (Fig. 2A. c). A ratio equal to one is sometimes attributed to dissolution of NaCl while a Na^{+}:Cl^{-} ratio greater than one might reflect release of Na^{+} from silicate weathering (Meyback, 1987). A significant correlation (r=0.60) exists between Na^{+} and Cl^{-} suggesting that they might be originating from the same source (Table 7). The increase in Na^{+} concentration was not associated with an increase in Cl^{-} concentration that supports a cation exchange process (Wayland et al., 2003). The plot of Na-Cl (Fig. 2A. c) also show that some samples deviate from the expected 1:1 relation indicating that a fraction of Na is associated with another anion. In the study area, the ratio of $Na^{+}/Cl^{-} < 1$ meaning another source is contributing chloride to the groundwater. Generally water containing significant amount of chloride whose drainage has few salty rocks or evaporate, is considered to be derived from the atmosphere (Al Mikhlafi et al., 2003). Among the anions, HCO_3^{-} and Cl^{-} are often dominant anions in groundwater. Precipitation of salts can cause a decline in the concentration of HCO_3^{-} and thereby anions in groundwater are dominated by Cl^{-}. The precipitation of $CaCO_3$ might result in a decline in Ca^{2+} concentration, supporting our finding of higher concentrations of Na^{+} in groundwater. As a result, the ratio of Mg^{2+}:Ca^{2+} was greater than unity in the groundwater showed in Fig. 2A (e). The sources of major cations, such as Ca^{2+} and Mg^{2+}, in groundwater can be the weathering of calcium and magnesium minerals (Krishna Kumar et al., 2009). In the areas of increased clay-rich soil dispersed and where Na^{+} concentration is higher (Yousaf et al., 1987), the Mg^{2+} concentration is relatively higher than that of Ca^{2+}. The Na^{+} hazard can be greater, where the groundwater is dominated by $Na^{+} + Mg^{2+}$ ions rather $Na^{+} + Ca^{2+}$ ions (Yousaf et al., 1987). The higher concentrations of Na decrease the hydraulic conductivity of soil/weathered materials, thus reducing the drainage conditions. The ratio HCO_3^{-}: Na^{+} can also be used to assess the weathering process (Krishna Kumar et al., 2009; Suba Rao, 2008) that occurs in groundwater. When the HCO_3^{-}: Na^{+} ratio is greater than 1, carbonate weathering occurs, while a ratio less than 1 allows the conclusion that silicate weathering occurs. In this study, all the groundwater samples had a ratio of Ca^{2+}: $HCO_3^{-} + CO_3^{2-}$ and Mg^{2+}: $HCO_3^{-} + CO_3^{2-}$ greater than unity while the ratio of Na^{+}: $HCO_3^{-} + CO_3^{2-}$ were approximately unity suggesting the predominance of Ca and Mg-containing minerals over Na-containing minerals in the study area. As a result, the ratios of $Ca^{2+} + Mg^{2+}$: total cations of most of the water samples had ratios approaching unity while the ratios of $Na^{+} + K^{+}$: total cations were far below unity (Fig. 2A).

Gibbs (1970) proposed a diagram to understand the relationship of the chemical components of waters from their respective aquifer lithologies. Three distinct fields, namely precipitation dominance, evaporation dominance, and rock dominance areas, are shown in the Gibbs diagram (Fig. 4). Gibbs ratios for the study area samples are plotted against their respective total dissolved solids as shown in Figure 4 to know whether the ground water chemistry is due to rock dominance, evaporation dominance or precipitation dominance. In the present study, Gibbs ratio observed from the diagrams fall into the precipitation dominance area.

Because our study area has a higher rate of evapotranspiration characterized by tropical climate and restricted fresh water exchange, salt layers may form near the evaporating surface (Karanth, 1994). On the other hand, human activities in areas of groundwater withdrawal might lead to future changes in groundwater chemical composition. For instance, intensive and long-term irrigation can leach salts from the soil/weathered rock zone, because of the availability of water.

Hydrogeochemical plots

The Na^{+} /Cl^{-} molar ratio in most of ground waters samples of the study area is less than 1 (Fig. 2A. c), indicating that silicate dissolution might not be a probable source for Na^{+} in groundwater of the aquifer. The groundwater derived from seawater intrusion will have Cl/Σanions greater than 0.8 and also have $Na^{+}/(Na^{+}+Cl^{-})$ ratio less than 0.5 (Hounslow, 1995). In this study, the average value of

Cl/Σanions ratio was lower than 0.8 and also Na^+/ (Na^++Cl^-) ratio was lower than 0.5 (Table 5). In addition, the simultaneous enrichment in both ions indicates dissolution of chloride salts or concentration by evaporation process (Jalali, 2005). This is responsible for the relatively high Na^+ and Cl^- contents in the saline groundwater. The higher concentration of Cl^- (mean value is 1.24 meq/L) explains the high degree of water/rock interaction in the form of silicate weathering. The plot of $Na^+ + K^+$ vs $Cl^- + SO_4^{2-}$(Fig. 2B. e) shows significant positive correlation. The decrease in alkalies $Na^+ + K^+$ with a simultaneous increase in $Cl^- + SO_4^{2-}$ does not reflect a common source for these ions. Sulfate ions would proceed from the oxidation of pyrite (FeS_2) as shown below:

$FeS_2+15/4\ O_2+7/2\ H_2O \rightarrow Fe(OH)_3 + 4\ H^+ + 2SO_4^{2-}$

This reaction would explain acidic nature of the groundwater. The decomposition of organic matter in the soil contributes also to the acidity of groundwater.

In case of carbonate weathering, the molar ratio of Ca^{2+} + Mg^{2+}/ HCO_3^- had greater than unity suggesting carbonate weathering is a dominant process and the source of high HCO_3^-. The plot of (Ca^{2+} + Mg^{2+}) vs (HCO_3^- + SO_4^{2-}) shows that most groundwater samples are plotted above the equiline (Fig. 2B. d). This indicates that carbonate weathering is the dominant process (Datta and Tyagi, 1996) for the supply of Ca^{2+} to the groundwater. The plot HCO_3^- vs Na^+ in (Fig. 2A. d) indicates that HCO_3^- ions are proceed from silicate and carbonate weathering. The plot of Ca^{2+}+Mg^{2+} versus total cations (TC), shows that the plotted points for all the samples fall in unity reflecting an increasing contribution of Ca^{2+} and Mg^{2+} (Fig. 2A. b). From the above, it is clear that the groundwater chemistry is likely to be from dissolution of silicate and carbonate minerals. Thus the concentrations of major elements are controlled by the congruent weathering of carbonate and incongruent weathering of silicates (Al-Mikhlafi, 2003; Garcia et al., 2001; Jalali, 2005). The ion exchange process is characterized by an (HCO_3^- + SO_4^{2-}) excess over (Ca^{2+}+Mg^{2+}), while the reverse ion exchange is marked by an excess of (Ca^{2+}+Mg^{2+}) over (HCO_3^- + SO_4^{2-}) (Fisher and Mulican, 1997). The ratio, $(Ca^{2+}+Mg^{2+})/(HCO_3^- + SO_4^{2-}) > 1$, indicates a reverse ion exchange process is contributing to the water chemistry in the study area (Fig. 2B. d).

By order of magnitude, carbonate weathering, silicate weathering and reverse ion exchange process control the groundwater composition in the study area. Sources of chloride in groundwater of the study area could be domestic wastewater. Taking in account the scatter diagrams in Fig. 2A and Fig. 2B, the carbonate weathering is the main hydrogeochemical process, followed by silicate weathering and reverse ion exchange process in the groundwater of the study area.

Suitability for irrigation

Parameters such as EC, pH, sodium sdsorption ratio (SAR), soluble sodium percentage (SSP) and Residual Sodium Carbonate (RSC) were used to assess the suitability of water for irrigation purposes (Ayers and Westcot, 1994; Islam et al., 2003; Hakim et al., 2009).

Suitability based on Ca^{2+}, Mg^{2+}, Na^+, K^+, PI and Kelly's ratio

The concentration of major cations such as Ca^{+2}, Mg^{+2}, Na^+ and K^+ in groundwater were far below the recommended maximum concentration of these parameters (20, 5, 40, 2 meq/L, respectively). The permeability index (PI) value is used to evaluate the sodium hazards of irrigation water and consequently is used as indicator whether or not the groundwater is suitable for irrigation. From the environmental point of view, a high permeability index, in association with subsurface structural features would facilitate widespread contamination of groundwater. Accordingly, waters can be classified as Class I, Class II and Class III orders. Class I and Class II waters are categorized as good for irrigation with 75% or more of maximum permeability. Class III waters are unsuitable with 25% of maximum permeability. The PI ranges from 12-66%, with average value is about 37% which locates under class-1 and class-2 (Table 4). The level of Na^+ measured against Ca^{+2} and Mg^{+2} is known as Kelly's ratio, based on which irrigation water can be rated. Groundwater having Kelly's ratio more than one is generally considered as unfit for irrigation. Kelley's ratio for the tested samples ranged from 0.03 and 0.56 indicating the suitability of all the samples for irrigation purpose.

Suitability for irrigation based on pH, EC, SAR and SSP

Irrigation water quality can have a profound impact on crop production. In this study, all of the samples were acidic in nature. Therefore, water with a low pH could contain elevated levels of toxic metals, cause premature damage to metal piping, and have associated aesthetic problems such as a metallic or sour taste, staining of laundry, and the characteristic blue-green staining of sinks and drains. All the groundwater samples might not be suitable for irrigation according to the results for pH since the acceptable pH for agricultural use ranges from 6.0 to 8.5 (Ayers and Westcot, 1985). We found significant variability in EC values, with the values ranging from 80 to 790 µS/cm. This result of EC was in close agreement in the groundwater of neighboring country Nepal reported by Chapagain et al. (2009). Higher EC values reflected higher concentrations of dissolved constituents that may affect the irrigation water quality in relation to salinity hazard. Table 6 shows that out of 44 samples, 40 were as 'excellent', 3 were rated as 'good' and 1 was as 'permissible' for irrigation purposes based on Wilcox requirement. The salts, besides affecting the growth of the plants directly, also affect soil structure, permeability and aeration, which indirectly affect plant growth (Singh et al., 2008).

Salts can accumulate in the root zone by two processes; the upward movement of a shallow saline-water table and salts left in the soil due to insufficient leaching. According to Richards (Richards, 1968), all irrigation waters were classified as C1S1 with nine C1S2, seven C1S3, one C1S4, one C2S1 and one C2S2 categories. C1 indicated 'low' salinity (EC < 250 μS/cm), C2 indicated 'medium' salinity (EC= 250-750 μS/cm), and S1 indicated 'low sodium', S2 indicated 'medium sodium' S3 indicated 'high sodium' and S4 indicated 'very high sodium' with respect to SAR. Irrigation with C1 and C2 class waters is unlikely to affect the osmotic pressure of the soil solution and the cell sap of the crop plants. If water with a high salinity level is used for a long period of time, the soil profile, especially the root zone, might be affected by the accumulation of salts which would ultimately reduce crop yields (Jahidul et al., 2010). The suitability of water for irrigation purpose depends on the sodium ion and total salt content of the water. Excess sodium in waters produces the undesirable effects of changing soil properties and reducing soil permeability. Plants intake water from soil by osmosis and osmotic pressure is proportional to the salt content, which affects the growth of plants, soil structure and permeability (Gupta et al., 2009). High concentrations of Na^+ are undesirable in water because Na^+ is adsorbed onto the soil cation exchange sites, causing soil aggregates to disperse, reducing its permeability (Jalali, 2009). The sodium adsorption ratio (SAR), which indicates the effect of relative cation concentration on Na^+ accumulation in the soil, is used for evaluating the sodicity of irrigation water. The degree to which irrigation water tends to enter into cation-exchange reactions in soil can be indicated by the sodium adsorption ratio. SAR is an important parameter for the determination of the suitability of irrigation water because it is responsible for the sodium hazard (Todd and Mays, 2005). With respect to the SAR values, all the groundwater samples were classified as excellent for crop irrigation and would not be expected to negatively affect soil quality (Table 6). Both a low salt content (low EC) and high SAR can mean there is a high potential for permeability or water infiltration problems. They can act separately or collectively to disperse soil aggregates, which in turn reduces the number of large pores in the soil. These large pores are responsible for aeration and drainage (Grattan, 2002). Therefore, some samples in the study area indicating very high salinity and very high sodium could be unsuitable for irrigation purposes.

Soluble sodium percentage (SSP) is an important criterion for soil physical properties and can affect plant growth. Among the groundwater samples we collected, 25 were rated as 'excellent', 12 were rated as 'good' and 1 was rated as 'poor' according to Wilcox. Water belonging to the 'excellent' and 'good' categories may be used for irrigation purposes. Natural occurrence of Na enriched minerals in parent materials and geochemical processes during interaction with groundwater may account for elevated Na concentration. As a result, soils may undergo deterioration due to the accumulation of Na, resulting in crust formation and seal development on the soil surface over a long period of time. Sodium toxicity is often modified and reduced if Ca and Mg are also present. Moderate amounts of Ca and Mg may reduce sodium damage and higher amounts even prevent it (Jahidul et al., 2010).

Suitability for irrigation based on TDS, RSC, H_T, Cl^-, NO_3^-, HCO_3^- and metals

The TDS values ranged from 54 to 529 mg/L with an average value of 123 mg/L. A water containing TDS less than 1000 mg/L can be considered to be 'fresh water' for irrigation use and will not affect the osmotic pressure of soil solution (Hakim et al., 2009). All the waters except one were rated as 'fresh' according to the guidelines given in Freeze and Cherry (1979). In our study, almost all the water samples were suitable for growing crops (Table 6). In addition to total dissolved solids, the relative abundance of sodium with respect to alkaline earths, and the quantity of bicarbonate and carbonate in excess of alkaline earths also influence the suitability of water for irrigation. This excess is denoted by 'Residual sodium carbonate' (RSC). The RSC is the most important criterion to examine the water quality. A negative RSC value indicates that the total concentration of CO_3^{2-} and HCO_3^- is lower than the sum of the Ca^{2+} and Mg^{2+} concentrations, reflecting that there is no residual carbonate to react with Na^+ to increase the Na hazard in the soil. As for the RSC values, all the water samples were graded as 'suitable' and these waters may be used safely. Natural concentrations of significant amounts of soluble Ca^{2+} and Mg^{2+} might be the possible cause of very hard water. Hardness in water is also derived from the solution of carbon dioxide released from the bacterial action in soil in percolating water. Among the samples, 6 samples were classified as 'soft', 31 samples were grouped as 'moderately hard', 5 samples were as 'hard' and 2 samples were 'very hard' waters (data not shown); these can be considered to be suitable for irrigation.

According to Ayers and Westcot (1985), the recommended concentration of chloride is 4.0 meq/L. Our results showed that chloride concentration ranged from 0.25 to 3.00 meq/L and all waters were suitable for irrigation. NO_3^--N concentrations in the groundwater samples ranged from 0.10 to 1.65 mg/L (Table 2). Our results showed that the groundwater NO_3^--N did not exceed the maximum concentration 30 mg/L as because most crops are relatively unaffected until N exceeds 30 mg/L (http://www.fao.org/docrep/003). Therefore, most crops can be irrigated in this region without any N hazard. Irrigation water containing CO_3^{2-} higher than 0.1 meq/L and HCO_3^- more than 10 meq/L are not generally recommended. All the samples are far below than the permissible limit. Therefore, all water samples in the study area can be used for long-term irrigation use (Jahidul et al., 2010).

Concentration of metals in groundwater including As, Fe, Mn, Cu and Zn is presented in Table 2. Most metal concentrations in water depending on the mineral solubility, and pH, Eh and salinity of the solution (Caron et al., 2008). Trace metals including Cu^{2+}, Zn^{2+}, Fe^{3+}, As^{3+}, Mn^{2+} were concentrations were low and considered to be suitable for crop production and the soil environment (Jahidul et al., 2010). However, the average concentrations of Cu^{2+}, Zn^{2+}, Fe^{3+}, As^{3+} and Mn^{2+} were 0.045 mg/L, 0.013 mg/L, 0.565 mg/L, 0.311 mg/L and 0.017 mg/L, respectively.

Correlations among the parameters with depth

Correlation coefficient is commonly used to establish the relation between independent and dependent variables. The correlation matrix of 14 parameters, for the 44 samples in the study area is indicated in Table 7. There were few significant relationships among the measured concentrations with depth. The high correlation between EC and TDS reflects the interdependency of these measurements as general measures of the amount of total dissolved solutes. The correlation between EC and TDS (r=0.96) is due to the fact that conductivity depends on total dissolved solids. The significant positive correlation (r=0.92) observed between SSP and SAR indicates that sodium adsorption by soil particles may increase with the amount of soluble sodium that may create an alkali hazard in soil hindering successful crop production. The high correlations between Cl^- and HCO_3^- (r=0.37), Cl^- and Na^+ (r=0.60), SO_4^{2-} and Mg^{2+} (r=0.66), K^+ and HCO_3^- (r=0.43), Na^+ and K^+ (r=0.39) and between Na^+ and HCO_3^- (r=0.55) indicating that they most likely derive from the same source of water (Table 3). The high positive correlation between Na^+-Cl^- may represent influence of saline water. The high positive correlation between Mg^{2+}-SO_4^{2-} and Ca^{2+}-SO_4^{2-} (r=0.45) may represent ion exchange and gypsum dissolution and the relation between Ca^{2+}-HCO_3^- (r=0.80) and Ca^{2+}-Na^+ (r =0.56) may represent contributions from silicate and carbonate weathering (Table 7). The correlation between K^+-NO_3^- (r=0.48) may represent poor sanitation conditions including application of fertilizer.

However, the relationship between EC and TDS indicates the increase of one element will or may increase the concentration of the other and synergistic behavior amongst the dissolved ions in water. Sunitha et al. (2005) identified that the EC finds higher level correlation significance with many of the water quality parameters, like TDS, chlorides, sulphates, total hardness and magnesium. Raman and Geetha (2005) found that the water quality of ground water can be predicted with sufficient accuracy just by the measurement of EC alone. This provides a means for easier and faster monitoring of water quality in a location.

Suitability for drinking and domestic uses

The analytical results of physical and chemical parameters of groundwater were compared with the standard guideline values as recommended by the World Health Organisation for drinking and public health purposes (WHO, 2004). In our survey, the pH (range 3.8 to 6.1) of all groundwater samples was not safe (safe limit 6.5-8.5) for drinking water prescribed by WHO (2004). The recommended concentration of TDS is 500 mg/L (WHO, 2004) and 2 samples were above the recommended limit. The degree of hardness in water is commonly based on the classification (0-75) soft, (75-150) moderately hard, (150-300) hard, (>300) very hard and hence groundwaters are soft (Todd and Mays, 2005). The maximum allowable limit of hardness for drinking water is 500 mg/L and the most desirable limit is 100 mg/L as recommended by WHO guidelines and all water were suitable for drinking. Hard water leads to high incidence of urolithiosis (WHO, 2004), anencephaly, parental mortality, some types of cancer (Agrawal and Jagetia, 1997) and cardio-vascular disorders. Such waters can develop scales in water heaters, distribution pipes and well pumps, boilers and cooking utensils, and require more soap for washing clothes (Todd and Mays, 2005; Karanth, 1994). In drinking water, excess concentration of some nutrients can cause health hazard. For example, the concentration of Na^+ should not exceed 200 mg/L. A sodium-restricted diet is recommended to patients suffering from hypertension or congenial heart diseases and also from kidney problems. For such people, extra intake of Na^+ through drinking water may prove critical. All groundwater samples have lower K^+ content than the acceptable limits (200 mg/L) for drinking water (WHO, 2004). The low levels of K^+ in natural waters are a consequence of its tendency to be fixed by clay minerals and to participate in the formation of secondary minerals (Pazand et al., 2011). Excess concentration of Cl^- gives a salty taste and has a laxative effect in people not accustomed to it. Higher concentration of SO_4^{2-} in drinking water is associated with respiratory problems (Subba Rao, 1993). Sulphate is unstable if it exceeds the maximum allowable limit of 8.3 meq/L and causes a laxative effect on human system with the excess magnesium in groundwater (Subramani et al., 2005). Excess NO_3^- can cause methemoglobinemia, gastric cancer, birth malformations and hypertension. However, the concentrations of Na, Cl^- , SO_4^{2-} and NO_3^- of the studied groundwater samples were far below the recommended limits (Na^+= 200 mg/L, Cl^- =250 mg/L, SO_4^{2-}=150 mg/L, NO_3^-=10 mg/L) for drinking according to WHO (2004). As most of the study area is intensively irrigated, the fertilizers used for agriculture may be the source for the elevated concentration of nitrate in a few locations (Chandna et al., 2010). The content of As is below the WHO guideline for drinking water (<0.01 mg/L) (WHO, 2004) but below the Bangladesh limit (<0.05 mg/L) (data not shown). Concentration of Fe of almost all the samples is above the WHO permissible limit (<0.01 mg/L). The mean value of Mn is 0.565 mg/L in the study area and above WHO guideline for drinking water (<0.05 mg/L). These higher concentrations of Fe and Mn might be responsible for the low pH value in the study area.

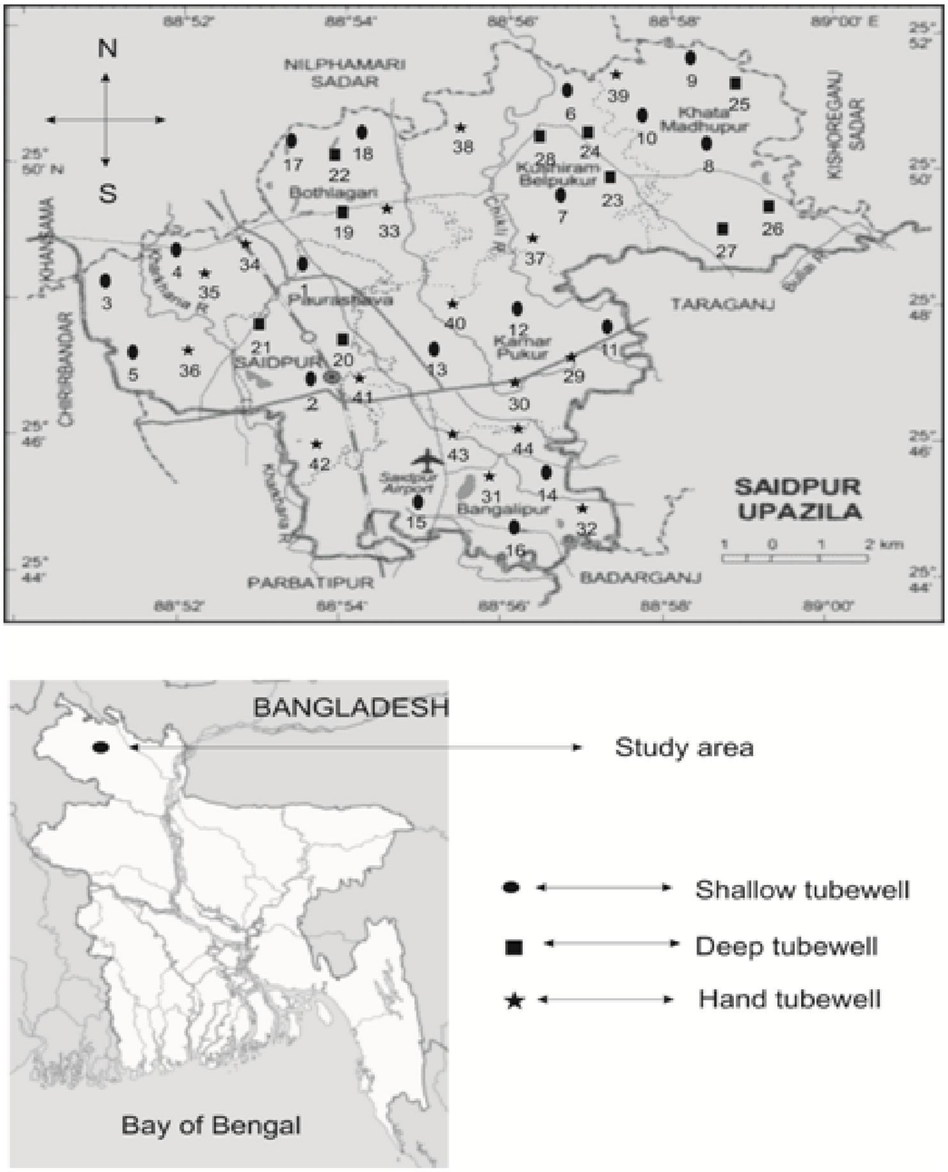
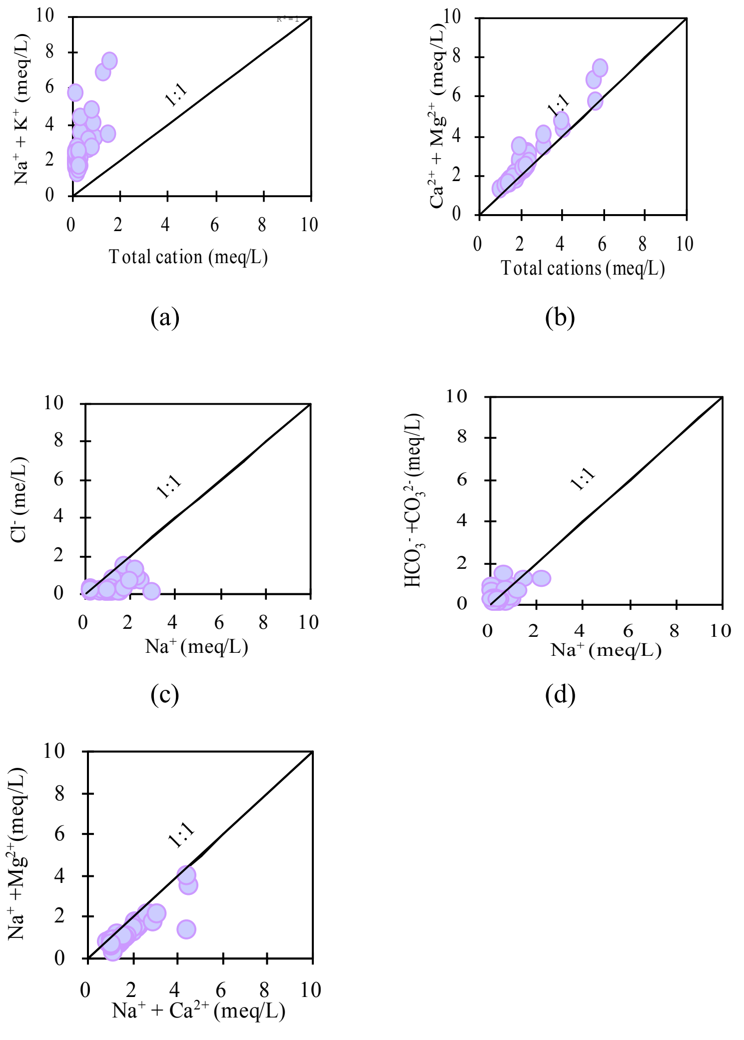


Fig. 1. Map of the sampling sites of the Saidpur Upazilla (16 shallow tubewell; 16 deep tubewell and 12 hand tubewell; total 44) under the District of Nilphamari along with the map of Bangladesh.

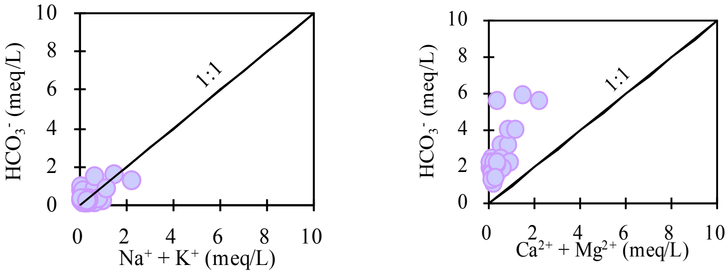


(a) (b)

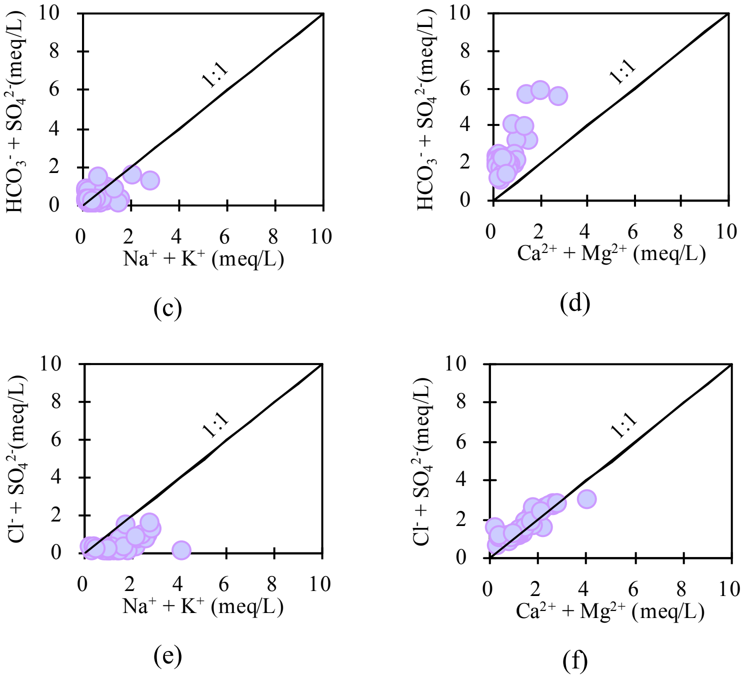
(c) (d)

(e)

Fig. 2A. Ratios of the major anions and cations in groundwater from Saidpur Upazilla, Bangladesh.



(a) (b)



(c) (d)

(e) (f)

Fig. 2B. Ratios of the major anions and cations in groundwater from Saidpur Upazilla, Bangladesh.

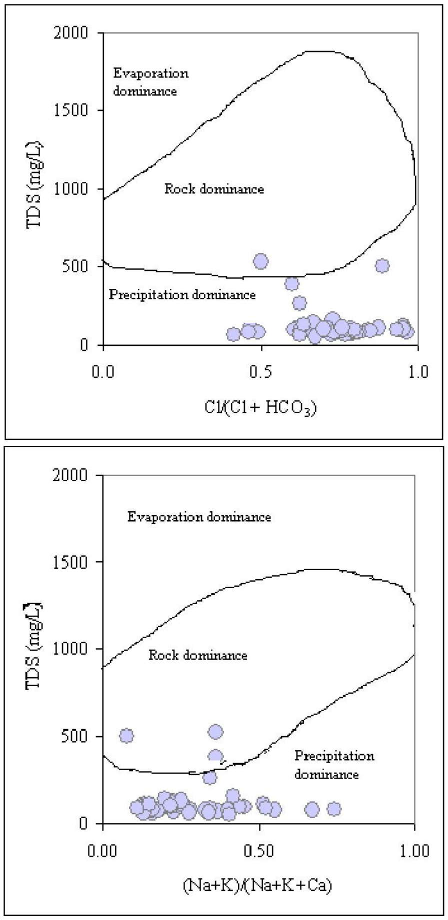


Fig. 3. Mechanisms controlling the quality of groundwater of Saidpur Upazilla (Gibbs, 1970)

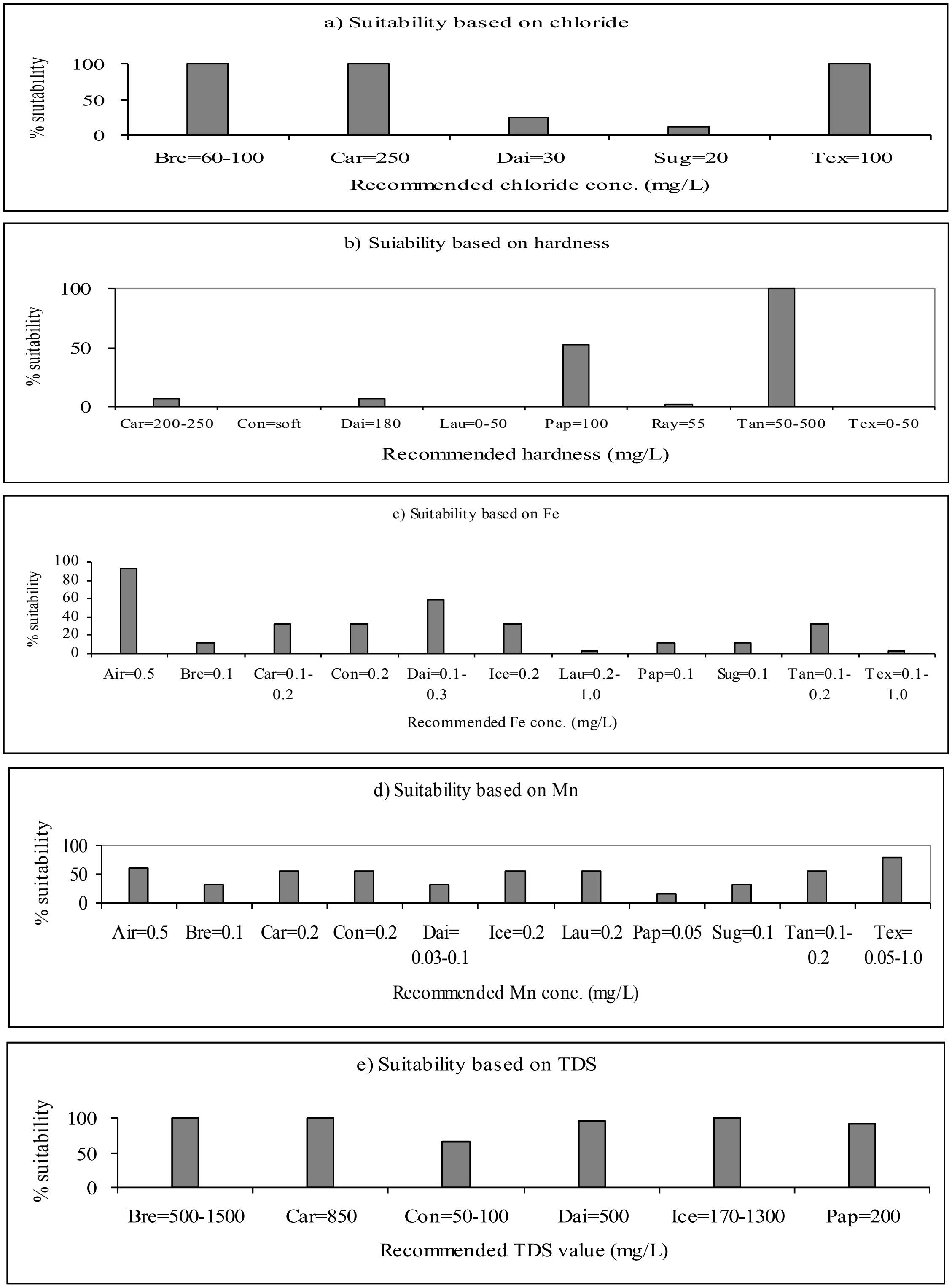


Fig. 4. Suitability of studied water samples for various industries based on a) hardness, b) chloride concentrations, c) Fe, d) Mn and e) TDS concentrations. In X axis, the recommended concentrations for different industries are shown according to Todd and Mays (2005). Air=Air-conditioning, Bre=Brewing, Car=Carbonated beverage, Con=Confectionary, Dai=Dairy, Ice=Ice manufacture, Lau=Laundering, Pap=Paper & pulp, Sug=Sugar, Tan=Tanning, Tex=Textile, Ray=Rayon manufacture

Table 1. Information regarding of sampling sites of the Saidpur Upazilla under the District of Nilphamari, Bangladesh; location, well type, depth, and the duration of uses

Sample	Sampling sites				
No.	Location	Union (Small administrative unit)	Depth of sink (m)	Well type	Duration of uses (year)
1	Phulpur	Pourashova	25	STW	10
2	Railbazar	Pourashova	25	STW	12
3	Kutipara	Bothlagari	45	STW	8
4	Dokhin mazapara	Bothlagari	20	STW	15
5	Teltola	Bothlagari	45	STW	14
6	Poshim belpukur	Kasiram belpukur	20	STW	16
7	Kasiram belpukur	Kasiram belpukur	30	STW	15
8	Khatamodhupur	Khatamodhupur	50	STW	17
9	Panisala	Khatamodhupur	12	STW	12
10	East Khatmodhupur	Khatamodhupur	22	STW	9
11	Bakdokra	Kamarpukur	25	STW	14
12	Kamarpukur	Kamarpukur	20	STW	12
13	Nijbari	Kamarpukur	28	STW	17
14	Laxmanpur	Bangalipur	45	STW	15
15	Cantonment	Bangalipur	30	STW	14
16	Dewanipara	Bangalipur	50	STW	12
17	Kisamot kadikhol	Bothlagari	16	STW	17
18	Bordoho	Bothlagari	18	STW	12
19	Dholipara	Bothlagari	80	DTW	14
20	New babupara	Pourashova	60	DTW	10
21	North Porashova	Pourashova	70	DTW	15
22	Khordobothlagari	Bothlagari	70	DTW	14
23	Poushim Belpukur	Kushiram Belpukur	65	DTW	18
24	Chowra	Kushiram Belpukur	68	DTW	20
25	Rotherpukur	Khatamodhupur	100	DTW	14
26	East Khatmodhupur	Khatamodhupur	85	DTW	19
27	South Khatamodhupur	Khatamodhupur	100	DTW	14
28	Central Kushiram	Kushiram Belpukur	75	DTW	22
29	Dolua	Kamarpukur	35	HTW	14
30	Niamotpur par	Kamarpukur	10	HTW	12
31	Central Bangalipur	Bangalalipur	30	HTW	10
32	South Bangalipur	Bangalalipur	13	HTW	11
33	Puraton porahat	Bothlagari	9	HTW	15
34	Bothlagari par	Bothlagari	25	HTW	14
35	Poraton porahat	Bothlagari	28	HTW	10
36	Togorpara	Bothlagari	40	HTW	8
37	Central Kushiram	Kushiram Belpukur	15	HTW	11
38	East Kushiram	Kushiram Belpukur	16	HTW	7
39	Hamurhat	Khatamodhupur	12	HTW	12
40	Aijdhal	Kamarpukur	25	HTW	14
41	Central Pourashova	Pourashova	10	HTW	13
42	West Pourashova	Pourashova	13	HTW	15
43	West Bangalipur	Bangalalipur	20	HTW	10
44	East Bangalipur	Bangalalipur	12	HTW	14

STW=Shallow tubewell, DTW= Deep tubewell, HTW= Hand tubewell

Table 2. pH, EC, TDS, hardness, and anionic constituents of groundwater during the study period

	pH	EC	TDS	Hardness	Cl^-	HCO_3^-	SO_4^{-2}	NO_3^-	PO_4^{3-}
		µS /cm	mg/L	mg/L	meq/L	meq/L	meq/L	mg/L	mg/L
Min	5.5	80	54	52	0.250	0.047	0.003	0.100	0.001
Max	7.5	790	529	428	3.00	2.25	1.08	1.65	0.66
Mean	6.4	192	123	120	1.24	0.45	0.19	0.54	0.10
SD	0.45	155	103	75	0.68	0.41	0.23	0.42	0.18

Table 3. Concentrations of cationic constituents of groundwater during the study period

	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	Cu^{2+}	Zn^{2+}	Mn^{2+}	Fe^{3+}	As^{5+}
	meq/L	meq/L	meq/L	meq/L	mg/L	mg/L	mg/L	mg/L	mg/L
Min	0.080	0.810	0.098	0.009	0.016	0.001	0.003	0.038	0.008
Max	2.73	4.37	1.45	0.33	0.065	0.030	2.250	2.620	0.032
Mean	0.85	1.46	0.36	0.04	0.045	0.013	0.565	0.311	0.017
SD	0.45	0.71	0.35	0.05	0.01	0.01	0.64	0.38	0.01

Table 4. Potential salinity (PS), permeability index (PI), Kelly's ratio, Gibbs ratio, sodium adsorption ratio (SAR) and residual sodium carbonate (RSC) of the groundwater

	PS	PI	Kelly's ratio	Gibbs ratio	Gibbs ratio	SAR	RSC
	meq/L			for anion	for cation		
Min	0.26	0.12	0.03	0.42	0.08	0.06	-5.50
Max	3.54	0.66	0.56	0.96	0.74	1.47	-0.85
Mean	1.33	0.37	0.20	0.73	0.29	0.33	-1.94
SD	0.73	0.10	0.12	0.14	0.16	0.30	1.01

Table 5. Stoichiometric ratios of different major ions in the studied water samples

Ratios	Min	Max	Mean	SD
$(Ca^{2+}+Mg^{2+})$/Tcations	0.57	0.99	0.86	0.09
$(Na^{+}+K^{+})$/Tcations	0.02	0.43	0.14	0.09
Ca^{2+}/Mg^{2+}	0.08	0.86	0.58	0.14
Na^{+}/Ca^{2+}	0.08	2.64	0.47	0.50
$Na^{+}/(Na^{+} + Cl^{-})$	0.03	0.56	0.22	0.13
HCO_3^{-}/Na^{+}	0.12	6.63	1.97	1.40
$(HCO_3^{-} + SO_4^{2-})$/Tanions	0.05	0.52	0.26	0.11
Cl^{-}/Tanions	0.13	0.87	0.51	0.19

Table 6. Quality classification of water samples for irrigation

Sl	EC	SSP	Water class based on		Alkalinity-
No	µS /cm		EC	SSP	salinity class
1	230	9	Ex	Ex	C1-S1
2	110	12	Ex	Ex	C1-S2
3	160	4	Ex	Ex	C1-S1
4	130	5	Ex	Ex	C1-S1
5	150	5	Ex	Ex	C1-S1
6	160	4	Ex	Ex	C1-S1
7	120	5	Ex	Ex	C1-S1
8	170	26	Ex	Good	C1-S3
9	120	25	Ex	Good	C1-S3
10	160	25	Ex	Good	C1-S3
11	190	28	Ex	Good	C1-S3
12	170	21	Ex	Good	C1-S3
13	240	22	Ex	Good	C1-S3
14	140	29	Ex	Good	C1-S3
15	130	5	Ex	Good	C1-S1
16	170	8	Ex	Good	C1-S1
17	120	6	Ex	Ex	C1-S1
18	150	9	Ex	Ex	C1-S1
19	170	9	Ex	Ex	C1-S1
20	120	6	Ex	Ex	C1-S1
21	120	6	Ex	Ex	C1-S1
22	140	5	Ex	Good	C1-S1
23	130	5	Ex	Ex	C1-S1
24	120	6	Ex	Ex	C1-S1
25	130	11	Ex	Ex	C1-S2
26	150	9	Ex	Ex	C1-S1
27	150	10	Ex	Ex	C1-S2
28	130	15	Ex	Ex	C1-S2

29	120	16	Ex	Ex	C1-S2
30	120	16	Ex	Ex	C1-S2
31	140	12	Ex	Ex	C1-S2
32	130	42	Ex	Poor	C1-S4
33	790	18	Per	Ex	C3-S2
34	750	2	Good	Ex	C2-S1
35	160	4	Ex	Good	C1-S1
36	160	4	Ex	Good	C1-S1
37	110	11	Ex	Ex	C1-S2
38	230	7	Ex	Ex	C1-S1
39	210	9	Ex	Ex	C1-S1
40	80	14	Ex	Ex	C1-S2
41	630	17	Good	Ex	C2-S2
42	390	14	Good	Ex	C2-S2
43	160	8	Ex	Ex	C1-S1
44	100	13	Ex	Ex	C1-S2

Ex = Excellent, Per=Permissible

Table 7. Correlation matrix of different chemical constituents of groundwater, n=44, units of each parameter are in Table 2 and Table 3

	Depth	EC	TDS	Cl^-	HCO_3^-	SO_4^{2-}	NO_3^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	SAR	SSP	H_T
Depth	1.00													
EC	-0.28	1.00												
TDS	-0.26	*1.00*	1.00											
Cl^-	-0.28	*0.61*	*0.60*	1.00										
HCO_3^-	-0.13	*0.73*	*0.72*	*0.37*	1.00									
SO_4^{2-}	-0.24	*0.66*	*0.64*	*0.35*	*0.29*	1.00								
NO_3^-	-0.24	*0.34*	*0.33*	-0.14	0.18	0.15	1.00							
Ca^{2+}	-0.32	*0.82*	*0.80*	*0.59*	*0.80*	*0.45*	*0.45*	1.00						
Mg^{2+}	-0.26	*0.93*	*0.93*	*0.70*	*0.62*	*0.66*	*0.36*	*0.79*	1.00					
Na^+	*-0.35*	*0.43*	*0.41*	*0.60*	*0.55*	0.15	0.11	*0.56*	*0.37*	1.00				
K^+	*-0.39*	*0.44*	*0.40*	0.17	*0.43*	0.21	*0.48*	*0.62*	*0.34*	*0.39*	1.00			
SAR	*-0.31*	0.20	0.18	*0.50*	*0.33*	0.03	0.01	*0.30*	0.17	*0.95*	0.21	1.00		
SSP	-0.25	0.01	0.01	*0.37*	0.14	-0.06	-0.09	0.07	-0.02	*0.86*	0.09	*0.96*	1.00	
H_T	-0.27	*0.97*	*0.97*	*0.67*	*0.67*	*0.67*	*0.34*	*0.81*	*0.98*	*0.39*	*0.32*	0.17	-0.02	1.00

Italic values are significant at $p = 0.05$ where $r \geq 0.29$

Industrial ratings

Based on pH (3.8 to 6.1), these waters might not be suitable for all industries (Todd and Mays, 2005). The TDS concentrations ranged from 54 to 529 mg/L. These concentrations were found suitable for brewing, carbonated beverage and ice manufacture. However, for confectionary, dairy, paper and pulp, the percent suitability were 66, 95 and 91, respectively (Fig. 4). Hardness was due to the sufficiency of divalent cations like Ca^{2+} and Mg^{2+} (Todd and Mays, 2005). As regards to hardness, almost all waters were unsuitable for confectionary, textile and laundering while the suitability were for tanning (100%), paper and pulp (52%), carbonated beverage (7%), rayon manufacture(2%) and dairy (7%) in the studied samples. Based on chloride concentration, the percent suitability for brewing, carbonated beverage, dairy, sugar and textile industries were 100,100, 25, 12 and 100, respectively. The allowable limits of Mn for various industries range from 0.05 to 1.0 mg/L except for sugar manufacture (Todd and Mays, 2005). For sugar manufacture water should be free from Mn. The concentration of Fe in the present study ranged from 0.038 to 2.620 mg/L and the groundwater sampled in this study might not be suitable for nearly all industrial uses with few exceptions.

Conclusion

The major cations in the studied groundwater is in the decreasing order as $Mg^{2+} > Ca^{2+} > Na^+ > K^+$. The anions are also arranged in decreasing order as $Cl^- > HCO_3^- > NO_3^- > SO_4^{2-}$, respectively. As because irrigation is a primary use of groundwater in this region, irrigation return-flow could become a cause of groundwater pollution in this region. If salts accumulate on the surface in irrigated fields, they can leach through the soil zone by recharge water and reach the water table. Further, recycling of groundwater for irrigation can result in a progressive increase in soil and groundwater salinity. Based on the patterns we observed, it can be concluded that all the hand tube well, shallow tube well and deep tube well water samples of the Saidpur Upazilla in the district of

Nilphamari, Bangladesh were suitable for irrigation, drinking, domestic and industrial uses; although some samples were rated to be unsuitable for some specific industries for some specific ions. Spatial variability is caused by aquifer heterogeneities, non-uniform agricultural loading patterns, and runoff of agricultural chemicals to topographically low points in the landscape.

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