BRAC University Journal, Vol. XI, No. 2, 2016, pp. 37-51

DETERMINING SOURCES of GROUNDWATER SALINITY IN THE MULTI-LAYERED AQUIFER SYSTEM OF THE BENGAL DELTA, BANGLADESH

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ABSTRACT

The study area lies on the coastal belt of the Bengal Delta, southern Bangladesh. This study has been conducted to determine salinity sources in the multi-layered aquifers of the Bengal Delta. Dissolved major ions including chloride (Cl^-) and bromide (Br^-) were measured in groundwater of the coastal areas of Bangladesh to assess the source(s) of salinity. Ionic concentrations of 48 groundwater samples were analyzed for this study; samples were collected from observation wells installed at different depths down to 336 m. The major ion trends of the upper (<200 m deep) Sodium (Na^+)-Chloride (Cl^-) groundwater type is defined as Na^+> Calcium (Ca^{2+})> Magnesium (Mg^{2+})> Potassium (K^+) and Cl^-> Bi-carbonate (HCO_3^-)> Sulfate (SO_4^{2-}). The deep groundwater (201-336 m deep) has ionic trend of $Na^+>Ca^{2+}>Mg^{2+}>K^+$ and $HCO_3^->Cl^->SO_4^{2-}$. The shallow groundwater (<200 m deep) of the south central coast has $Br^-:Cl^-$ ratios between 0.00234 and 0.00354 and corresponding $Cl^-:Br^-$ molar ratios between 635.51 and 959.78, which indicates that seawater from the ocean is the principal source of chloride in the upper aquifers with the mixture of fresh water mainly recharged from monsoon rain. At some locations in the south-central coast, seawater is also the principal source of the salinity in deep groundwater, within the depth ranging from 201 to 336 m. Elevated chloride concentration might be due to the entrapment of relict seawater in the sediment during Holocene transgression.

Keywords: Groundwater salinity, coastal plain, sources of salinity, bromide and chloride ratio.

Introduction

Bangladesh lies mostly in the Bengal delta that is characterized by its complex drainage and river system. The major portion of the delta is crisscrossed by many large and small channels, some of which are losing their flow due to drying up and siltation; some are active and others disperse water only during high tides. This drainage pattern also plays an important role in the saline water distribution of both surface water and the aquifer system in the southern coastal belt of the country. The coastal area covers about 20% of Bangladesh and over 30% of the net cultivable area within19 southern districts. The area lies on active, inactive and tidal delta and differs from the rest of the country because of its complex lithologic, hydrogeological and geochemical characteristics. Increased use of groundwater in Bangladesh since 1969 led to its wider use as the major source of potable and irrigation water supply. Pumping of groundwater accelerates salt water intrusion and degradation of water quality along both horizontal and vertical salinization paths. Water salinity creates ecological and socio-economic problems by limiting availability of fresh water resources and restricting crop production that ultimately could hamper food security of the country.

The aquifer formations of Bangladesh were developed by the fluvial and estuarine unconsolidated sediments of Pleistocene to Recent age. Currently, about 80% and 98% of irrigation and drinking water supply respectively for the country is provided by groundwater. But, this water resource is facing threats in many areas due to pollution from agriculture and contamination of arsenic in shallow groundwater, which makes the water harmful for drinking purpose (BWDB 2013; Bhattacharya et al. 1997; DPHE-BGS 2001; Nickson et al. 1998; Ravenscroft et al. 2001; van Geen et al. 2003; Yan et al. 2000; Zahid et al. 2009a, 2008). Salinization especially in the coastal aquifers and eco-system is another threat to groundwater quality (BWDB 2013; Zahid et al. 2013). The coastal area of Bangladesh is densely populated, consisting of single-family homes serviced by individual shallow and deep wells tapping the aquifer. Fresh water resources are considered valuable assets in the coastal zone where the surface water is saline most of the year. The coastal population is suffering from the lack of available fresh, safe water due to salinity in the upper aquifer as well as in deep groundwater in many places (down to the depths of 250-300 m). High salinity in groundwater limits its use for domestic, agricultural and industrial applications.

Groundwater salinity is commonly described by the chloride content, total dissolved solids content (TDS), or electric conductivity (EC) of the water. Saltwater intrusion may cause high chloride concentrations in groundwater exceeding the WHO (2008) and U.S. Environmental Protection Agency (1986) allowable limit of 250 mg/l for chloride. Chloride concentrations between 21 and 19,133 mg/l have been detected in groundwater in the coastal areas of southern Bangladesh as part of this research. Understanding the hydraulic and hydrogeochemical conditions and determining the source(s) of salinity is important to assessing the influence of seawater encroachment on coastal groundwater (Mahesha and Nagaraja 1996). Historically, absolute and relative concentrations of specific solutes like sodium, chloride, bromide etc. were frequently used to perform regional and/or local scale hydrochemical interpretation. No major hydrogeochemical assessment has been conducted to date in Bangladesh to detect the source(s) of salinity in coastal groundwater. Dissolved chloride is the most dominant chemical constituent in seawater. Bromide, which is chemically similar to chloride, is also present in seawater at low concentrations. The average concentrations of chloride and bromide in sea water are generally 19,000 and 65 mg/l, respectively (Hem 1992). Typical concentrations of chloride and bromide in fresh (non-saline) groundwater are lower, i.e. less than 10 mg/l for chloride and below reporting level for bromide. Chloride and bromide do not take part in environmental chemical reactions because of their chemically conservative nature in aqueous settings (Andreasen and Fleck 1997). As a result, the ratio between bromide and chloride ($Br^-:Cl^-$) and chloride and bromide ($Cl^-:Br^-$) is generally constant with time and across locations. If seawater mixes with fresh water, the concentrations of chloride and bromide decrease with increasing distance from the sea and depth in the water column, but the $Br^-:Cl^-$ and $Cl^-:Br^-$ ratio remains constant. Previous studies depict that $Br^-:Cl^-$ and $Cl^-:Br^-$ molar ratios in groundwater intruded with sea water are similar to those in seawater (Alcala and Custodio 2008; Richter and Kreitler 1993). The $Br^-:Cl^-$ in Bay water is equal to the $Br^-:Cl^-$ in seawater, i.e. 0.0033 to 0.0037, if dilution is considered as the only process acting on dissolved chloride and bromide (Yongje Kim et al. 2003; Hem 1992; Morris and Riley 1966). Many authors also indicate that relatively uniform chloride and bromide concentrations occur in the major reservoir of water (the ocean) and their $Cl^-:Br^-$ molar ratio is about 655 ± 4 (Fontes et al. 1986; Whittemore 1988; Davis et al. 1998; Custodio and Herrera 2000). Analytical error in the chemical analysis may account for the difference between the ratios.

The principal objective of this paper is to determine the source(s) of groundwater salinity in the multi-layered coastal aquifers down to the depth of about 350 m using concentrations of major ions in groundwater and $Br^-:Cl^-$ ratios.

Sampling and Methodology

The Bangladesh Water Development Board (BWDB 2013) has established a monitoring network to assess water resources potential in the 19 coastal districts of southern Bangladesh. Under this network, 42 groundwater monitoring well nests have been installed at 42 locations (Figure 1). Each nest consists of 3 to 5 piezometers at different depth levels down to the maximum depth of 350 m; the nest are used for measuring the water table, collection and physio-chemical analysis of water samples and performing different hydraulic tests. Amongst 48 analyzed groundwater samples for this study, sample nos.16, 14, 11 and 7 were collected from aquifer depths of less than 100, 101 to 200, 201 to 300, and 301 to 336 m, respectively. At a few locations aquifer units of all or many of these different depth ranges are separated by impermeable or leaky aquitards, while in other locations no significant aquitard is encountered throughout the aquifer depth until a sampling depth of 350 m. Lithologic logs were interpreted to map the extent of aquifer units as the coastal lithology of Bengal Delta is highly variable and complex; this complexity plays a role in the mobility and uneven distribution of salinity. Locations of the sampling wells are presented in Figure 1. Nine locations were selected in the south-central coast, where salinity concentration is comparatively higher; two locations lie on Cox's Bazar Coastal Plain, and one is from the Tippera Surface (GWTF 2002), where salinity intensity is lower. Groundwater samples were collected in dry season i.e. between November and March, 2012-2013.

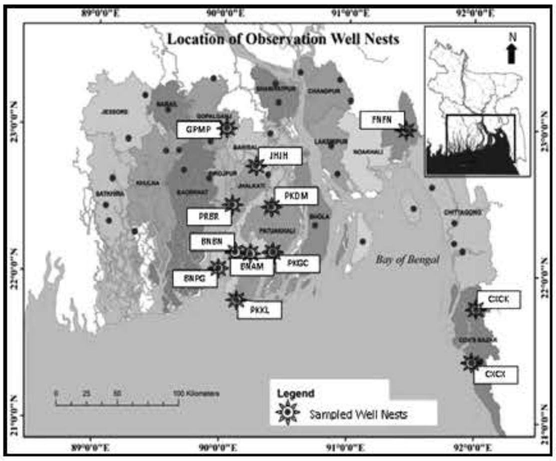


Fig. 1 - Location map of the sampling observation well nests (piezometers) in the deltaic coastal areas of Bangladesh. Each nest consists of 3 to 4 piezometers.

Dissolved chloride concentrations and electric conductivity (EC) of collected groundwater samples were measured to identify the areas affected by brackish and saline water intrusion, and to map the maximum extent of the fresh water/brackish water interface at different depths in the aquifers. Brackish water is defined as water with Chloride and EC concentrations of 300-600 mg/l and 1000-2000 µS/cm respectively (BWDB 2013). Samples having concentration above and below these limits are defined as saline and fresh water, respectively. The upper unconfined aquifers are susceptible to surficial contamination in addition to saline-water intrusion. Major cations such as Ca^{2+}, Mg^{2+}, Na^+ and K^+ and anions such as Cl^-, HCO_3^-, SO_4^{2-} and nitrate (NO_3^-) were also measured to classify the water types. To present and classify groundwater considering major ions in water and characterize the hydrochemical parameters between various sources of water Piper diagrams are used by many authors (Piper 1944; Soulsby et al. 1998).

The bromide and chloride ratios in water of the coastal aquifers were compared to $Br^-:Cl^-$ ratios and $Cl^-:Br^-$ molar ratios in sea water to determine the source of salinity. As both bromide and chloride ions are conservative chemically in undisturbed aqueous environments (i.e. they are non-reactive in redox reactions, neither do they sorb onto surfaces of mineral or organic substances nor form insoluble precipitates), therefore, these are good indicators of salinity (Fetter 1993). High Cl^- concentrations, $Br^-:Cl^-$ ratios of groundwater within the normal seawater range of 0.0033 to 0.0037, and a mean $Br^-:Cl^-$ ratios of 0.0036 indicates that the salinization in groundwater is caused mainly by the mixing of seawater (Yongje Kim et al. 2003). Where dilution is the lone process acting on dissolved chloride and bromide in the Bay, the $Br^-:Cl^-$ in Bay water is generally equal to the $Br^-:Cl^-$ in seawater, i.e. 0.0034 to 0.0035 (Hem 1992; Morris and Riley 1966). Sea water is the source of the chloride in groundwater if $Br-:Cl^-$ ratios in groundwater and seawater are similar. Differences in ratios indicate that at least a portion of chloride is derived from another source of chloride (Andreasen and Fleck 1997). The chloride and bromide concentrations in seawater are uniform, at 655 ± 4 for the $Cl^-:Br^-$ molar ratio (Fontes et al. 1986; Whittemore 1988; Davis et al. 1998). This ratio may slightly vary depending on analytical accuracy and local effects. To determine the groundwater and surface water salinity origin and source, the $Cl^-:Br^-$ ratio has been used as a tracer (Rittenhouse 1967; Carpenter 1978; Freeman 2007), and is an established and useful tool in hydrogeological investigations with low-to-moderate salinity of surface and groundwater (Whittemore 1988; Davis et al. 1998; Vengosh and Hendry 2001; Cartwright et al. 2006). Nitrogen (as nitrate) concentrations were analyzed to determine if the source of chloride was from sewage effluent. Elevated chloride concentrations can also be sourced from the contamination of groundwater by fertilizer nitrate (Richter and Kreitler 1993).

Atomic Absorption Spectrometry (AAS) was used to measure cations, while anions, including chloride, were determined by using ion chromatography (IC). Bromide was measured by using colorimetry. The groundwater samples were analyzed in the laboratory of the Department of Geology, University of Dhaka. EC values were measured in the field during sampling using EC meter.

Results and Discussion

Aquifer System in the Coastal Area

In the coastal areas of the Bengal Delta, hydrogeology as well as distribution of the type of aquifer sediments in the subsurface is very complex (BWDB 2013; Zahid et al. 2014). Aquifer-aquitard alteration is highly variable even within a short distance. The Meghna Flood Plain, Chittagong-Cox's Bazar Coastal Plain and the Ganges-Brahmaputra-Meghna Delta Plain dominate the coastal area. Acquired data from several thousands of bore holes was used to define the extent of aquifer formations. The depositional rate of the sediment and subsidence in the Bengal Delta were not uniform during the Quaternary period. As such, at similar depths, sediments of different depositional environments and various ages can be detected. Terminology of aquifer units have been defined by many studies. BWDB-UNDP (1982) described three aquifers, on regional basis. Considering isotopic studies and depth of aquifers, Aggarwal et al. (2000) classified four types of groundwater and proposed a three-tier division of the aquifer units. In coastal areas these aquifers can be classified as (BWDB 2013; BWDB-UNDP 1982), (i) the shallow or the 1st aquifer, (ii) the main or the 2nd or the lower shallow aquifer, and (iii) the deep or the 3rd aquifer. The shallow, i.e. the 1st aquifer, the upper Holocene aquifer, that extends down to 50 to over 100 m, in many places below a considerably thick aquitard of clay and silt unit. The aquifer sediments consist of fine sand with lenses of clay. Aggarwal et al. (2000) and Zahid et al. (2014) mention it as the First Aquifer. Based on groundwater isotope and model studies, average age of water of this aquifer is dated between 40 and 135 years (Aggarwal et al. 2000; Zahid et al. 2014). The extension of the main or the 2nd water bearing zone is down to 200-250 m and is confined by silty clay aquitards composed predominantly of fine to very fine sand, interbedded with clay lensesin places. These Mid-Holocene Aquifers may be considered as the Main Aquifer (BWDB-UNDP 1982) or the 2nd Aquifer or the Lower Shallow Aquifer (BGS-DPHE 2001). The average age of water from this floodplain and deltaic aquifers is dated as about 2,300 to 3,000 years old (Aggarwal et al. 2000; Zahid et al. 2014). The deep, i.e. the 3rd aquifer, has been encountered to depths of 250-350 m, generally below an aquitard and consists mainly of grey to dark grey fine sand intercalated with thin silty clay or clay lenses. The appearance of aquitards is not common in all locations and aquifers down to the investigated depth of 350 m seem to be hydraulically connected on a regional basis. However, in many places 3 to 4 aquifer units are encountered separated by aquitards and limited scale abstraction of groundwater from any aquifer depth does not affect the others. The Late Pleistocene-Early Holocene Deep Aquifers mentioned in BWDB-UNDP study (1982), the lower part of the Deep Aquifer of the BGS-DPHE types (2001) and the 3rd Aquifer as defined by Aggarwal et al. (2000) and Zahid et al. (2014) are dated as about 1,400 to 20,000 years old, depending on the depth variations, geology and hydraulic characteristics of the aquifer formations.

In the sampling locations of the south-central delta, the eastern coastal plain and the Tippera surface spatial and vertical distribution of aquifer-aquitard sediments are very complex with varied depths and thickness of these units (Figure 2). The numbers

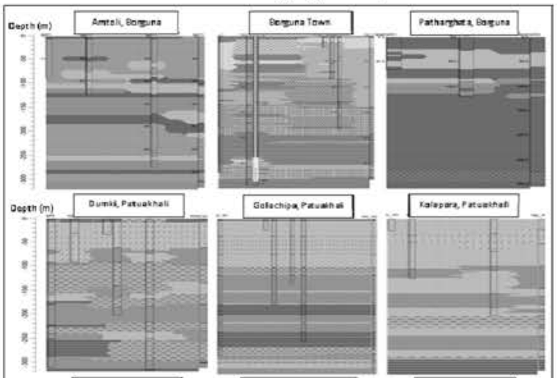


Fig. 2: Lithologic cross-sections of the sampling aquifers. Screens of wells with the length of 3 m are installed in the bottom of piezometers above a bail plug of 3 m ling.

and extent of aquitards investigated at depths to 350 m at different locations are highly variable; this influences uneven distribution and variations of salinity in groundwater both spatially and vertically. The erratic occurrence of small fresh water pockets at depth overlain and underlain by saline waters is reported all over the coastal belt. The thick clay layers are leaky or even absent in some places, so it does not provide the closed conduit necessary for flushing saline aquifers. The deep aquifer in many areas is confined by an overlying clay sequence which protects the fresh water from saline water intrusions, except near the estuaries.

The groundwater in the area occurs in inter-granular porosity found primarily in the basin filled alluvial sediments. Groundwater with similar chemical signature generally has similar hydrologic characteristics, similar recharge mechanism and flow paths in terms of climate, mineralogy, and age of water and formation sediment (Guler et al. 2002).

Hydraulic conductivity values of the multi-layered aquifer sediments, estimated by conducting slug tests, ranges between 1 and 25, 1 and 9 and 1 and 9 m/day for the shallow, the main and the deep aquifers, respectively, which is typical for sandy alluvial aquifers (Zahid et al. 2013) and transmissivity were estimated between 100-2300, 100-2200 and 100-1600 m^2/day, respectively, for the different depth levels of aquifer sediments. Transmissivity values of the deep aquifers, estimated from long duration (up to 72 hours) aquifer pump tests, depict that the Jhalokathi, and Patuakhali deep aquifers have higher potential with transmissivity ranges between 769 and 3224 m^2/day, while the Barguna, Barishal and Cox's Bazar deep aquifers show moderate potential with transmissivity values between 493 and 916 m^2/day. The Feni deep aquifer shows low potential with transmissivity ranges between 144 and 370 m^2/day. Storage co-efficient values of the deep aquifers were estimated between 0.0044 and 0.00016, indicating that the deep aquifers are leaky confined to confine in nature, overlain by leaky to impermeable clay aquitards.

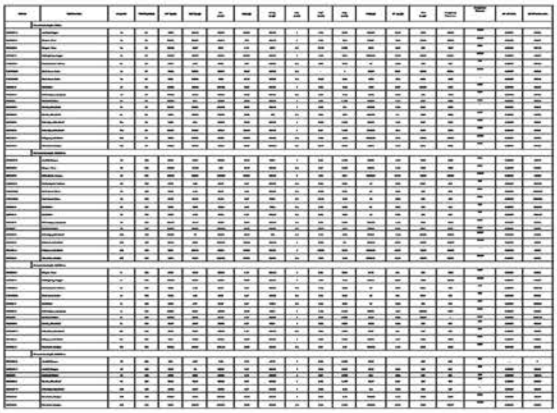
Groundwater Type

Mineral constituents and grain size of the aquifer sediment, composition of precipitation, climate, topography, etc. influences the chemical composition of groundwater. These factors control diverse water types that change both spatially and vertically. In the study areas of the coastal delta, very fine sand and silt dominate the aquifer sediment. Fine-grained sediments allow very slow percolation of water flow through pore spaces. Clay, silt, and fine sand facilitate dissolution of minerals because of long exposure periods. The most common exchange reactions are the water softening reactions when groundwater moves through clay-rich sediment where Ca^{2+} and Mg^{2+} in the water exchange with sorbed Na^+ (Appelo and Postma 1999). The monovalent ions are replaced by the divalent ions that are generally strongly bonded. However, at high activities, divalent ions can be replaced by the monovalent ions (Fetter 1994). Saline water encroachment from the Bay and through rivers and salt-rich precipitation near the coast can also play a vital role in groundwater chemistry.

Major ions in groundwater

Analytical results of the major ions of 48 groundwater samples of coastal aquifers, collected from the depth ranges of 24 to 335 m of the multilayered aquifers, are presented in Table 1. Respired CO^2 from the degradation of oxic and anoxic organic matter accelerates processes of weathering which further reacts with carbonate and silicate minerals to release Ca^{2+}, Mg^{2+}, Na^+ and K^+ into the groundwater (Zheng et al. 2004). The major cation concentrations for the depth ranges of <100, 101-200, 201-300 and 301-336 m are, Ca^{2+} (47.97-632.95, 14.87-407.1, 14.23-275.62 and 13.02-285.5 mg/l), Mg^{2+} (9.56-1092.8, 6.46-803.24, 4.79-105.04 and 4.42-121.76 mg/l), Na^+ (43.03-8531.5, 30.76-7487.45, 23.99-2204.9 and 6.98-2773.54 mg/l) and K^+ (3.97-320.63, 1.73-283.75, 2.84-33.99 and 2.18-18.77 mg/l), respectively. The deeper fresh groundwater has lower concentrations for all measured cations. Sources of Na+ and Cl^- influence the presence of higher proportions of Na^+ in comparison to Ca^{2+}, Mg^{2+}, and K^+ and high Cl^- in groundwater (Zheng et al. 2004). Recharging shallow groundwater is generally contaminated by Cl^- sourced from fertilizer in agricultural application of K^+ (Bohlke 2002), in addition to the seawater intrusion in the coastal belt. Assimilation into growing plants and ion exchange with clay minerals play an important role in determining the concentration level of K^+ in water. In agricultural recharge, enrichment of agricultural K^+ over background concentrations is supported by K^+:Cl^- ratios which are generally less than 1 (Bohlke 2002).

Table 1. Major ions and bromide-chloride ratios in dry season groundwater of the multi-layered aquifers in the coastal areas of the Bengal Delta



The concentrations of major anions for the depth ranges of <100, 101-200, 201-300 and 301-336 m are, HCO_3^- (167.75-1235.25, 205.88-648.13, 144.88-686.25 and 49.75-503.25 mg/l), CO_3^- (0-4.5, 0-3.0, 0-4.5 and 1.5-3.0 mg/l), SO_4^{2-} (0.04-81.36, 0.09-189.7, 0.09-8.54 and 0-0.89 mg/l) and NO_3^- (0-46.3, 0.02-40.8, 0.06-18.88 and 0.336-5.55 mg/l) respectively. The deeper i.e. fresh groundwater has lower concentrations for all measured anions. SO_4^{2-} is found to be higher (up to 468.75 mg/l) in shallow samples. NO_3^- concentrations (upto 46.3 mg/l) are also higher in few shallow samples. Recharging agricultural NO_3^- reacts with sulfide minerals of aquifer sediment to form SO_4^{2-} by denitrification (Van Beek et al. 1989). $SO4^{2-}$ can be retarded by sorption and added to fields with NH_4^- or Ca^{2+}. Reduction of SO_4^{2-} generally causes very low SO_4^{2-}/Cl^- ratios and is not due to low SO_4^{2-} with recharging water (Zheng et al. 2004).

The large variations of total dissolved ions are most apparent for Na^+ (6.98-8531.5 mg/l), Cl^- (21-19132.85 mg/l) and HCO_3^- (49.75-1235.25 mg/l) concentrations. Presence of dissolved Cl^- is common in shallow groundwater, but natural concentrations are generally low (Hem 1992). Except in the Feni and Cox's Bazar area, groundwater Cl^- concentrations generally fall within salinity ranges (Cl> 600 mg/l) for the upper groundwater samples down to 200 m depths. Below this depth the deep groundwater has Cl^- concentrations mostly within fresh water limit of 300 mg/l. Presence of dissolved HCO_3^- concentrations in groundwater support water-rock interaction and microbial degradation of organic substances (Zheng et al. 2004). The weathering of calcite or silicate minerals originates the secondary mineral formation by atmospheric or respired CO^2 groundwater alkalinity or HCO_3^- (Garrels 1967).

Groundwater classification based on Piper diagram

To define the groundwater types in an aquifer the hydrochemical facies that differ in their chemical composition are used (Fetter 1994). The lithology, solution kinetics, and flow patterns of the aquifer influences formation of facies (Back 1960, 1966). The Piper diagram (Piper 1944) is the trilinear graphical diagram that is commonly used to classify groundwater. On two separate trilinear plots the Piper diagram presents the relative concentrations of the major cations and anions and on a central diamond plot the points from the two trilinear plots are projected. The quadrilateral central diamond-shaped field is used to present overall chemical character of groundwater (Hill 1940; Piper 1944). The subdivisions of the diamond field represent water-type categories for natural waters (Back 1961; Back and Hanshaw 1965). The mixing of water from different sources including mixing of fresh water and seawater or evolution pathways can also be determined by this diagram (Freeze and Cherry 1979). Groundwater from acoastal aquifer may show a surplus of Ca^{2+} which indicates seawater intrusion, or an enrichment of Na+ which indicates fresh water intrusion (Appelo and Postma 1999). The conservative mixing is presented by the plot in between fresh and seawater compositions in the Piper diagram.

Figure 3 presents the results the 48 groundwater samples on the Piper diagram. Employing the water classification scheme of Fetter (1994) and Back (1961) and average concentrations in fresh water and seawater (Appelo and Postma 1999) using the Piper trilinear diagram, the chemical compositions of the analyzed samples are described for this study. Almost all of the groundwater samples down to the depth of 200 m, except Cox's Bazar and Feni samples, are distributed in the right central portion of the diagram and are classified as Cl^--SO_4^{2-}, Cl^--SO_4^{2-}-HCO_3^-, Na^+-Ca^{2-}, Na^+-K^+ types, dominated by Na^+-Cl^- and HCO_3^- type. Seawater intrusion is the major phenomena up to this depth. Deep water (down to 336 m) of Patharghata, Bhandaria and Mukshedpur aquifers is also classified with this cluster. Cox's Bazar and Feni samples from down to 200 m depth shows Na^+-Ca^{2+} and HCO_3^--Cl^- SO_4^{2-} types, with the signature of the mixture of fresh water and seawater. Most of the deep groundwater samples between 201 and 336 m depth ranges are classified as HCO_3^--Cl^--SO_4^{2-}, Na^+-K^+-Ca^{2+} types, dominated by HCO_3^- type; these are within the limits of fresh water. The main ion trends of the upper Na^+-Cl^- groundwater are $Na^+>Ca^{2+}>Mg^{2+}>K^+$ and $Cl^->HCO_3>SO42^-$ and for the deep groundwater ionic trend is $Na^+>Ca^{2+}>Mg^{2+}>K^+$ and $HCO_3>Cl^->SO_4^{2-}$.

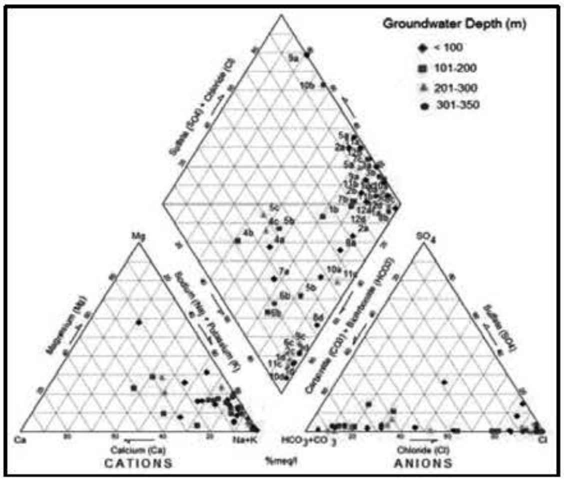


Fig. 3: Piper diagram showing groundwater types of multi-layered aquifers in the Bengal Delta.

Distribution of Salinity

In coastal aquifers salinity is the most prominent form of groundwater degradation which is characterized by the enrichment of total dissolved solids (TDS), Electric Conductivity (EC) and chemical constituents like Cl^-, Na^+, Mg^{2+}, and SO_4^{2-} (BWDB 2013). Sources of groundwater salinity are diverse. Common sources include: natural saline groundwater, presence of paleo-brackish water, halite dissolution, intrusion of seawater, and agricultural and industrial effluents. Seawater encroachment is the most widespread source of salinity in coastal aquifers. The present fresh water-saline water interface is the limit of potable water from the deep aquifers mainly in the coastal areas of Bangladesh, and is fairly well-defined. It can be seen that the fresh water-saline water interface lies about 75 to 100 km inland from the coast line in upper aquifers with the variable intensity of salinity (Figure 4).

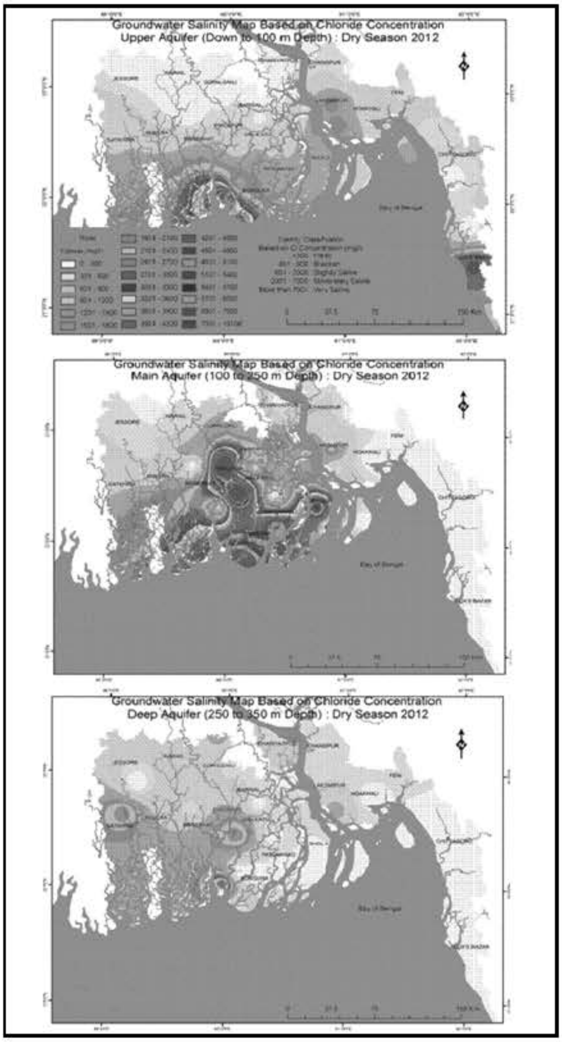


Fig. 4: Distribution of dry season groundwater chloride concentrations at different depth levels in the coastal aquifers of the Bengal delta.

The occurrences of brackish and saline water in the coastal aquifers do not follow any regular pattern spatially or vertically. All depth levels of aquifer units investigated down to 336 m have been affected by salinity in many areas (Figure 4). In the coastal delta, salinity of the upper composite and main aquifers is extremely variable and changes abruptly over short distances. In most of the study areas, the water is too saline for domestic and irrigation use due either to seawater encroachment or estuarine flooding. In some areas, flushing out of the saline water has resulted in fresh water pockets. The change from potable water to very saline water is sharp and occurs over a relatively short distance. Figure 4 shows contour maps of groundwater chloride concentrations at different depth levels of coastal aquifers based on BWDB data (BWDB 2013). The maps show that in the shallow aquifer (<100 m depth) fresh to brackish groundwater ($Cl^- <300$ mg/l) is noticed in areas of Borgunasadar, Mukshedpur and Chokoria, while brackish water (Cl^-=300-600 mg/l) occurs at Golachipa area. Salinity ($Cl^- >600$ mg/l) occurs in the shallow aquifers of the rest of the study areas. Fresh water occurs in the upper part (101-200 m deep) of the main aquifers of Feni, Cox's Bazar sadar and Chokoria areas. Brackish water is detected in Amtoli aquifer. Groundwater in the upper part of the main aquifers of the rest of the areas of the study area contains moderate to high salinity with the chloride content up to 15,970 mg/l. In the lower part of the main aquifer (201-300 m deep) fresh groundwater occurs at Borgunasadar, Amtoli, Feni, Cox's Bazar, Dumki and Golachipa areas, while brackish water is detected at Mukshedpur and Kolapara. Groundwater of Patharghata and Bhandaria areas at this depth levels contain salinity with the chloride content up to 4,367 mg/l. Except for Bhandaria, deep groundwater (300-336 m deep) remains fresh in all investigated aquifers under study area.

Seasonal variability of salinity (EC) is also noticed and generally, dry season groundwater salinity is higher than wet season salinity in all aquifer units of the coastal delta (Table 1). However, this variation is significant in shallow groundwater and low in deep groundwater. In the upper aquifers, to depths of 100 m, the salinity is extremely variable, overlain by very shallow fresh water pockets recharged from recent precipitation that changes rapidly over short distances. Groundwater EC values vary from 31 to 43,300 and 39 to 44,600 µS/cm in dry and wet seasons, respectively, in the 1st aquifer. The maximum EC value of dry and wet seasons were measured at 43,300 and 44,600 µS/cm, respectively, at Patharghata. In the 2nd aquifer, the EC ranges from 309 to 37,300 µS/cm in dry season, and in the wet season ranges from 109 to 34,100 µS/cm. Maximum EC values for the dry season and wet season were detected as 37,300 and 34,100 µS/cm, respectively, at Patharghata. The deep or 3rd aquifer shows groundwater EC values between 231 and 37,000 µS/cm in dry season and 72 and 10,340 µS/cm in wet season. A maximum EC value of 37,000 µS/cm was observed in the dry season at Kolapara, and maximum value of 10,340 µS/cm was observed in the wet season at Bhandaria. Seasonal variations of EC values for each location are presented in Table 1.

Source of Salinity

To evaluate seawater encroachment on coastal aquifers, it is important to understand the salinity source(s) and to determine the hydraulic connectivity between different aquifer units, as well as hydrogeochemical conditions. The Cl^- and Br^- ratio and other ionic concentrations of groundwater samples have been used in this study as indicators to analyze the origin of groundwater salinity.

Generally, the sources of higher chloride concentrations of groundwater samples, except few samples of Cox's Bazar and Feni aquifers, is mainly due to mixtures of fresh water and seawater, as chloride concentration of most of the samples exceed 50-5,000 mg/l (Yongje Kim et al. 2003). The TDS and major ion, e.g., Na^+ and Cl^-, relationships in the groundwater were plotted to determine the major parameters that influence groundwater quality (Figure 5). The plot, involving samples from all depths up to 336 m, depicts that these ions have positive correlations with TDS. There is a strong correlation of Na^+ and Cl^- with TDS (r^2 =0.9175 to 0.9908 and r^2 =0.8469 to 0.9765, respectively) depicting that these ions have originated from the same saline water source (Zahid et al. 2008). Na^+ shows a strong correlation with Cl^- (r^2 =0.8299 to 0.9956) also, indicating that the same source of saline waters is the origin of these ions. The fresh groundwater and seawater mixing is also supported by the relationship between $Cl^-:HCO_3^-$ ratios and Cl^-. Few water samples show lower ratios of $Cl^-:HCO_3^-$ and Cl^- and can be considered as fresh water.

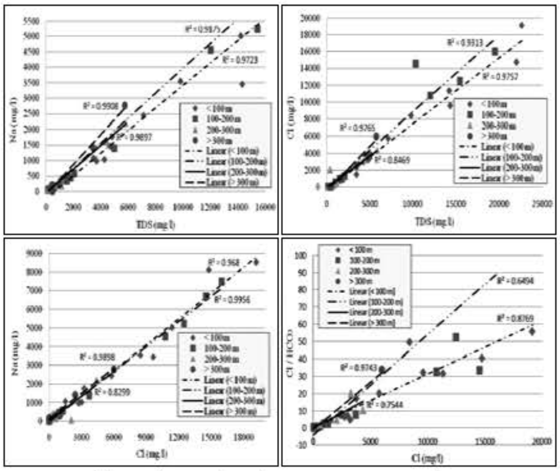


Fig. 5: The relationship between TDS and major ions (e.g. Na^+ and Cl^-) in the groundwater of the multi-layered coastal aquifers of the Bengal delta.

The presence of chloride and sulfate in deep groundwater and within the confined aquifers generally reveals that higher chloride and sulfate concentrations in these aquifer units were due to the encroachment of seawater during the last eustatic high (Amleto and Pucci 1999), thousands of years ago.

Bromide and Chloride ratio to differentiate salinity source

The absolute concentrations of bromide and chloride can be changed by the physical processes, e.g., dilution, evaporation, transpiration, etc. taking place in soil, but these processes cannot modify the $Br^-:Cl^-$ or $Cl^-:Br^-$ molar ratio of moderately saline groundwater significantly (Alcala and Custodio 2008). The $Br^-:Cl^-$ ratios and corresponding chloride concentrations are plotted in Figure 6; the $Br^-:Cl^-$ or $Cl^-:Br^-$ molar ratio values are presented in Table 1. Two trend lines are included for comparison in the plot. One line is the theoretical $Br^-:Cl^-$ ratio, equivalent to the $Br^-:Cl^-$ ratio in seawater (Hem 1992), and the other characterizes groundwater with background bromide concentration affected by a chloride source not containing bromide (Andreasen and Fleck 1997). This trend line is meant to represent anthropogenic chloride-affected native groundwater.

The occurrence of $Br^-:Cl^-$ ratios that are similar to the Bay water ratio in groundwater samples (i.e. 0.0033–0.0037 or $Cl^-:Br^-$ molar ratio of 655 ± 4) indicate that seawater is the primary source of the chloride (Andreasen and Fleck 1997), which may be variable slightly depending on analytical accuracy and local effects. The shallow groundwater (<100 m deep) of the south central coastal Delta (i.e. beneath Amtoli, Patharghata, Borgunasadar, Mukshedpur, Jhalokathi, Dumki, Golachipa, Kolapara and Bhandaria) has $Br^-:Cl^-$ ratios between 0.00234 and 0.00354 and corresponding $Cl^-:Br^-$ molar ratio-s between 636and 960. Groundwater within the depth ranges of 101 to 200 m under Amtoli, Patharghata, BorgunaSsadar, Mukshedpur, Jhalokathi, Dumki, Golachipa, Kolapara and Bhandaria aquifers has $Br^-:Cl^-$ ratios between 0.00246 and 0.00316 and corresponding $Cl^-:Br^-$ molar rations between 712and 870, indicating that seawater from the. Bay of Bengal is the primary source of chloride/salinity in the upper aquifers in these areas, with the mixture of fresh water from streams and recharged precipitated water. The seawater portion is very high in Patharghata, Jhalokathi, Dumki, Golachipa and Kolapara shallow groundwater with $Br^-:Cl^-$ and $Cl^-:Br^-$ molar ratios between 0.00302 and 0.00354 and 636and 745. Chokoria, Cox's Bazar sadar and Feni groundwater has $Br^-:Cl^-$ or $Cl^-:Br^-$ molar ratios ranges from 0.00598 to 0.00897 and 250to 293 and 0.00025 to 0.00061 and 4733to 11946 for <100 and 101-200 m, respectively. These concentrations are dissimilar from that of Bay water, and this groundwater does not have elevated nitrogen (as nitrate) concentrations (<3.00 mg/l), indicating fresh water (Andreasen and Fleck 1997).

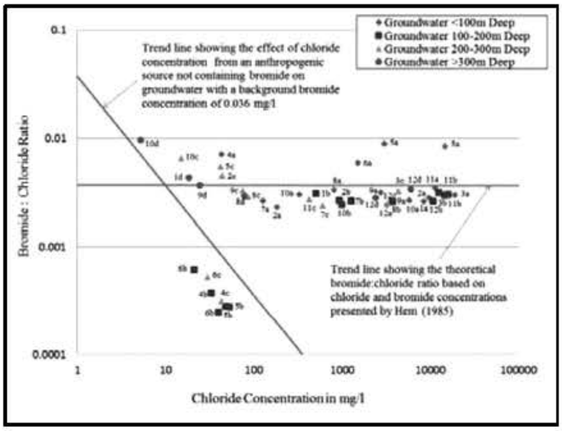


Fig. 6: Bromide: Chloride ratios related to chloride concentrations in groundwater of the multi-layered coastal aquifers of the Bengal Delta.

In the unsaturated zone, recharging water has a short residence time, and the addition of non-atmospherically derived sources of chloride and bromide to groundwater are negligible. The expected groundwater $Cl^-:Br^-$ ratios can be controlled by the salinity in atmospheric bulk deposition. Increased salinity due to evapoconcentration does not change groundwater $Cl^-:Br^-$ ratios during recharge (Fontes and Matray 1993; Davis et al. 1998; Edmunds 2001).

Deep groundwater within the depth ranges of 201 to 336 m under the Patharghata, Mukshedpur, Jhalokathi, Dumki, Kolapara and Bhandaria aquifers has $Br^-:Cl^-$ ratios between 0.00243 and 0.00367 and corresponding $Cl^-:Br^-$ molar rations between 613and 882 indicating that seawater from the Bay of Bengal is the major source of chloride/salinity in the deep aquifers in these areas. The elevated chloride concentrations mainly in deeper confined aquifers of the Bengal Delta may be due to relict seawater entrapped in the sediment during the Holocene transgression. As seawater mixes with fresh water, the concentrations of chloride and bromide decrease with increasing distance from the mouth of the Bay and depth in the water column, but the $Br^-:Cl^-$ ratio remains constant. With increasing distance from the sea, atmospheric chloride and bromide availability decreases. Therefore, chloride and bromide deposition rates are higher near the coast than inland (Eriksson 1960; Davis et al. 1998; Edmunds 2001; Alcala′ and Custodio 2008). Deep groundwater of Chokoria, Cox's Bazar sadar, Feni and Golachipa aquifers contain fresh water with $Br^-:Cl^-$ ratio ranges from 0.00032 to 0.00961 and is dissimilar from that of Bay water.

With elevated concentrations of chloride, $Cl^-:Br^-$ ratios in water can rapidly be increased due to halite dissolution (Kloppmann et al. 2001). The soil organic matter can generally adsorb both chloride and bromide ions (Gerritse and George 1988; O¨berg and Sanden 2005) and thus the $Cl^-:Br^-$ ratio changes in water. $Cl^-:Br^-$ ratio can also be reduced because of release of bromide during deforestation and soil organic matter is oxidized by ploughing (Alcala and Custodio 2008). In both coastal and inland areas, vegetation cover and industrial activities can contribute natural and anthropogenic atmospheric dust and organic molecules (Yvon-Lewis and Butler 2002) that modify the $Cl^-:Br^-$ ratios of bulk deposition (Martens et al. 1973; Alcala′ and Custodio 2005). Studies have shown that the $Cl^-:Br^-$ ratios increase between 1,200 and 7,500 in groundwater, although $Cl^-:Br^-$ ratios up to 10,000 have been documented (McCaffrey et al. 1987; Cartwright et al. 2004), due to the dissolution of halite containing sulfate-rich evaporates. The $Cl^-:Br^-$ ratios of groundwater in the upper aquifers of Cox's Bazar sadar and Feni are 10819.16 to 11946.16 and 9015.97,respectively. The groundwater of these aquifers is also enriched with sulfate concentrations ranges from 35.6 to 45.12 mg/l and 9.62 mg/l, respectively. Due to the density difference between fresh and saline water, the shallow brackish and saline water can penetrate deep into the aquifer layers, contaminating the fresh water, except where the deep fresh water is protected by more or less continuous clay layers.

The southwestern and south-central portion of the Bengal Delta, covering the Sundarbans, the world's largest mangrove forest, is a huge network of tidal creeks and channels. A substantial quantity of water flows through its various distributaries which join these tidal channels and estuarine creeks. Shifting of courses is a common characteristic of these rivers. The delta behaves as a fluvial delta during the monsoon season when about 3 million cusec (cubic feet per second) of water passes through the drainage network, whereas in the winter it behaves as a tide dominated delta when the volume of water passing through the channels reduces to 250,000 to 300,000 cusec. These uncommon features make this delta one the most complex on the Earth and influences the distribution and concentration levels of salinity with regard to space and depth. For the upper aquifers, salinity is also related to the relative amounts of saline and fresh water flooding from estuarine tidal effects.

Lithologic logs and seasonal groundwater table fluctuation trends in the alluvial and fluvio-deltaic multilayered aquifers of the Bengal delta supports hydraulic connectivity of the aquifer units at different depth levels down to 350 m depth regionally (Zahid et al. 2009b). In the studied multi-layered aquifers where significant clay aquitards exist, groundwater of different aquifer units has no hydraulic connectivity, at least locally, and the depth to groundwater tables are different (Figure 2 and Figure 7). In general, deeper aquifers have a higher groundwater level (above mean sea level) in natural conditions, but the seasonal fluctuation trend of the groundwater table from piezometers installed at different depths at a particular location is similar. However, as the deep production wells (>200 m deep) are the main source of municipal water supply in the coastal towns, groundwater level in many deep aquifers like studied aquifer under Borguna town, have lower head than that of the upper aquifers due to over exploitation of groundwater. Therefore, lithology, i.e. hydraulic properties of aquifer sediments, and development stresses of groundwater abstraction are playing a very important role to influence flowpaths and travel time of salinity transport in groundwater.

The lithologic variability (Figure 2) and trend of groundwater table fluctuations (Figure 7) at different depth levels in the studied aquifers have influenced the mobility of saline water. At Borguna district, the deep aquifer is overlain by the silty clay aquitard at Borguna town and Amtoli locations and is separated from the upper aquifers. The deep groundwater in the area is fresh, with the Cl^- concentration ranging between 10 and 43 mg/l in comparison to the water salinity of the aquifers above. Conversely, at Patharghata area, about 15-

20 km west of Borguna town, although a 150+ m thick aquitard separates the deep aquifer from the upper aquifers, the Cl^- concentration remains high in the deep groundwater, with a value of about 4,367 mg/l. The $Br^-:Cl^-$ ratios depict that moderate to high groundwater salinity in the upper aquifers in the area is due to the encroachment of the sea-water. Entrapped sea-water might be the source of Cl^- at Patharghata deep aquifer, as the thick clay aquitard retards the mobility of recent saline water from the aquifers above. The significant difference of groundwater levels between the deep and the upper aquifers in these three upazila areas also supports the hydraulic separation of the deep and the upper aquifers. At Patuakhali the deep aquifer is separated from upper aquifers by clay and siltyclay aquitards, and the deep groundwater is generally fresh in all studied aquifers (Cl^- concentration ranges between 5 and 423 mg/l). At Kolapara, deep groundwater has higher Cl^- concentrations (423 mg/l) in comparison to the other areas; this might be due to the leakage of saline water from the upper aquifers through the relatively thin silty clay aquitard and aquitard windows. The static groundwater levels in both the upper and deeper aquifers are very close during monsoon except for the Dumki aquifer, where the deep water table is lower than the water tables in the upper aquifers.

No significant impermeable clay is encountered between upper and deep aquifer units at Muksedpur and Bhandaria area and saline groundwater occurs throughout the investigated depth of 350 m. The deep groundwater in these two areas is classified as brackish and with low salinity, with Cl^- concentrations levels of 600 and 2,400 mg/l, respectively. The $Br^-:Cl^-$ ratios indicate that sea water is the source of salinity in the aquifer system. The trends of groundwater levels also depict the hydraulic connectivity between the deep and the upper aquifers. The deep groundwater in Feni and Cox's Bazar areas are classified as fresh, although the shallow groundwater is saline, and no mixing of sea water in the deep groundwater is observed. Alternate layers of silty clay aquitards that overlie the deep aquifer in Feni area may retard the leakage of saline water from above, and the higher head and pressure of recharged water from the hilly areas may push the saline water towards the sea in Cox's Bazar area. The deep groundwater in the Jhalokathi area is also fresh, with the Cl^- concentration ranging between 78 and 85 mg/l. However, there is evident signature of mixing of the seawater with the deep groundwater, like the groundwater described above.

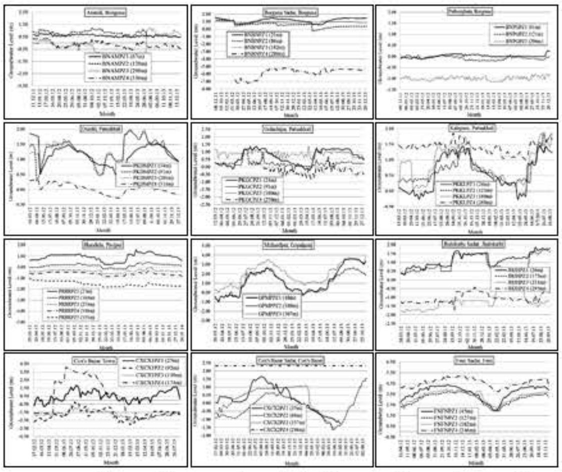


Fig. 7: Groundwater level hydrographs of sampling observation well nests installed at different depth levels.

Aquifers in areas with lower topographic relief, i.e. south-central delta, are much more vulnerable to intrusion pathways than aquifers in higher-relief, i.e. southeastern delta areas. Pumping of fresh groundwater in the coastal aquifer accelerates saltwater intrusion and degradation of water quality along both horizontal and vertical salinization paths. This indicates a clear need for hydrogeologic characterization, management of pumping, and hydrologic and geochemical monitoring of the coastal aquifer in Bangladesh, irrespective of future climate change.

Any future sea-level rise will increase the adverse impact of the already-occurring salinization processes to some extent. Moreover, an increased frequency of storm surges, or storm surges that cover a greater area of the land surface due to the higher sea stand, will increase the likelihood of vertical downward intrusion of saltwater to wells that currently produce fresh groundwater, wherever the saline floodwater is able to infiltrate.

Conclusion

In the coastal areas of the Bengal Delta, i.e. the south-central delta, the eastern coastal plain and the Tippera surface, hydrogeology as well as spatial and vertical distribution of aquifer-aquitard sediments is very complex with varied depths, thickness and extension of these units. The numbers and extents of aquitards up to the investigated depth of 350 m at different locations are highly variable; that, along with development stresses of groundwater abstraction, influences uneven distribution and variations of salinity in groundwater both spatially and vertically. The large ranges of total dissolved ions have remarkable variations of Na^+ (6.98-8,532 mg/l), Cl^- (21-19,133mg/l) and HCO_3^- (49.75-1,235.25 mg/l) concentrations. The groundwater Cl^- concentrations are generally high and above the limit of fresh to brackish water (Cl> 600 mg/l) for the upper groundwater samples down to 200 m depths in the south-central coast. Below this depth, deep groundwater has Cl^- concentrations mostly within fresh water limit of 300 mg/l or below. Almost all of the groundwater samples down to a depth of 200 m (except the eastern coastal plain and the Tippera surface samples), are classified as Cl^--SO_4^{2-}, Cl^--SO_4^{2-}-HCO_3^-, Na^+-Ca^{2+}, Na^+-K^+ types, dominated by Na^+-Cl^- and $HCO3^-$ type. The eastern coastal plain and the Tippera surface samples down to a 200 m depth indicate- Na^+-Ca^{2+} and HCO_3^--Cl^--SO_4^{2-} water types. Most of the deep groundwater samples between 201 and 336 m depth ranges are classified as HCO_3^--Cl^--SO_4^{2-}, N^+-K^+-Ca^{2+} types, dominated by HCO_3^- type; these are within the limit of fresh water levels. The major ion trends of the upper Na^+-Cl^+ groundwater type are: $Na^+>Ca^{2+}>Mg^{2+}>K^+$ and $Cl>HCO_3>SO_4^{2-}$, and for the deep groundwater ionic trend is: $Na^+>Ca^{2+}>Mg^{2+}>K^+$ and $HCO_3^->Cl>SO_4^{2-}$.

Besides natural saline groundwater, there are many different sources of salinity that may contaminate fresh groundwater like halite dissolution, presence of paleo-brackish water, seawater intrusion and domestic, agricultural and industrial effluents. The occurrences of brackish and saline water in the coastal aquifers do not follow uniform pattern spatially or vertically. All the different depth levels of aquifer units down to the maximum investigated depth of 336 m have been affected by salinity in many areas. In the coastal delta, salinity of the upper composite and main aquifers is extremely variable and changes abruptly over short distances.

Generally, the sources of higher chloride concentrations of groundwater samples in the study area can be considered as mixtures of fresh water and saline water except for a few samples of the eastern coastal plain and the Tippera surface aquifers. Na^+ and Cl^- show a strong correlation (r^2 =0.9175 to 0.9908 and r^2 =0.8469 to 0.9765, respectively) with TDS indicating that such ions are derived from the same source of saline water. In addition, Na+ shows a strong correlation with Cl^- (r^2 =0.8299 to 0.9956), indicating that these ions are also derived from the same source of saline water. The relationship between $Cl^-:HCO_3^-$ ratios and Cl^- shows mixing of fresh groundwater and seawater, and the samples can be characterized as fresh water with lower ratios. Chloride and bromide concentrations were detected between 21 and 19,133 and 0 and 133 mg/l, respectively, in the studied samples. The shallow groundwater (<200 m deep) of the south-central coastal delta has $Br^-:Cl^-$ ratios between 0.00234 and 0.00354 and corresponding $Cl^-:Br^-$ molar rations between 635.51 and 959.78 indicating that seawater from the Bay of Bengal is the primary source of chloride, i.e. salinity in the upper aquifers in these areas. At some locations in the south-central coast, seawater is also the major source of the salinity for deep groundwater within the depth range of 201 to 336 m. The occurrence of higher chloride and sulfate concentrations in deep groundwater and within the deeper confined aquifer might have happened due to seawater entrapped in the sediment during the Holocene transgression. Lithology, i.e. hydraulic properties of aquifer sediments, and development stresses of groundwater abstraction are playing a very important role in influencing flowpaths and travel time of salinity transport.

Acknowledgement

Bangladesh Water Development Board (BWDB), Bangladesh Climate Change Trust (BCCT) under the Ministry of Environment and Forest, and Institute of Water Modelling (IWM) are highly acknowledged for supporting the research under Climate Change Trust Fund project to assess coastal water resources. Farhana Tazneen, Geologist, BWDB, is acknowledged for preparing the chloride maps. Hydrogeologists of the project team are acknowledged for their valuable cooperation in the field sampling and laboratory activities.

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