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ORIGINAL ARTICLE

Hydrogeochemical evaluation and statistical analysis of groundwater of Sylhet, north-eastern Bangladesh

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Abstract To investigate the hydrogeochemical characteristics of groundwater 23 shallow, 30 intermediate and 38 deep wells samples were collected from Sylhet district of Bangladesh, and analyzed for temperature, pH, Eh, EC, DO, DOC, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, $SO_4{}^{2-}$, $NO_3{}^-$, $HCO_3{}^-$, $SiO_2{}^-$, Fe, Mn and As. Besides, 12 surface water samples from Surma and Kushiyara Rivers were also collected and analyzed to understand the influence into aquifers. Results revealed that, most of the groundwater samples are acidic in nature, and Na–HCO_3 is the dominant groundwater type. The mean value of temperature, EC, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, $NO_3{}^-$ and $SO_4{}^{2-}$ were found within the range of permissible limits, while most of the samples exceeds the allowable limits of Fe, Mn and As concentrations. However, relatively higher concentration of Fe and Mn were found in deep water samples and reverse trend was found in case of As. The mean concentrations of As in shallow, intermediate and deep wells were 39.3, 25.3 and 21.4 μg/L respectively, which varied from 0.03 to 148 μg/L. From spatial distribution, it was found that Fe, Mn and As concentrations are high but patchy in northern, north-western, and south-western part of Sylhet region. The most influential geochemical process in study area were identified as silicate weathering, characterized by active cation exchange process and carbonate weathering, which thereby can enhance the elemental concentrations in groundwater. Pearson's correlation matrix, principal component analysis and cluster analysis were also employed to evaluate the controlling factors, and it was found that, both natural and anthropogenic sources were influencing the groundwater chemistry of the aquifers. However, surface water has no significant role to contaminate the aquifers, rather geogenic factors affecting the trace elemental contamination. Thus it is expected that, outcomes of this study will provide useful insights for future groundwater monitoring and management of the study area.

Keywords Arsenic · Groundwater · Hydrogeochemistry · Multivariate statistics · Spatial distribution

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1 Introduction

Water is a finite commodity in the world, and used for drinking, agricultural and industrial needs. Urbanization, increase of population, dewatering of aquifers for irrigation and extensive use of domestic and industrial activities are common phenomenon, that have direct effects to deteriorate the water resources in different parts Bangladesh including the Sylhet region (Bhuiyan et al. 2016; Rahman et al. 2017; Islam et al. 2017a, b, 2018). The quality of water is of vital concern for mankind, as it is directly linked with human welfare (Kumar et al. 2018b). Poor quality of water adversely affects the plant growth and human health (Hem 1991; Islam et al. 2017c). Water quality gets modified along the course of movement through the hydrological cycle and the operation of the others processes including evaporation, transpiration, uptake by vegetation, oxidation/reduction, cation exchange, dissociation of minerals, precipitation of secondary minerals, mixing of waters, leaching of fertilizer sand manure, pollution and biological processes (Thilagavathi et al. 2012; Islam et al. 2017c; Wagh et al. 2017).

However, in Asia, over 1 billion people are directly dependent on groundwater resources (Foster 1995), but the quantity and quality is affected by many natural and anthropogenic causes (Bhuiyan et al. 2016; Kumar et al. 2018a; Sakram and Adimalla 2018). Former studied revealed that, nearly 97% people of Bangladesh are dependent on groundwater, where approximately 86% of the extracted water is used for irrigation (BADC 2002; Hasan et al. 2007). But, from the last few decades, different part of Bangladesh is facing limited groundwater resources with quality deterioration due to unplanned and over exploitation, industrial and agricultural activities and others natural processes (Bhuiyan et al. 2015; Bodrud-Doza et al. 2016; Islam et al. 2017c, 2018). Moreover, surface water is also an important source to meet the daily needs of inhabitants, which influenced by the climate variation, river shifting, river flow, urbanization, agricultural practices, domestic and industrial activities (Biswas et al. 2014; Rahman and Islam 2016; Islam et al. 2016). Furthermore, groundwater quality depends on the quality of recharge water from surface sources (Kumar et al. 2014). Besides, different hydro-geochemical processes also governing the chemical characteristics of groundwater, and well documented by many authors i.e., Thilagavathi et al. (2012), Sivasubramanian et al. (2013), Kumar et al. (2014), Bhuiyan et al. (2016), Islam et al. (2017c) and Islam et al. (2018). Thus the knowledge of hydrogeochemistry is essential to determine the origin of chemical composition of groundwater (Thilagavathi et al. 2012). Therefore, detailed investigations on the groundwater hydrogeochemistry and influence of surface water regarding quality deterioration are imperative. Halim et al. (2010a) and Islam et al. (2017a) reported about the arsenic contamination and groundwater quality degradation in eastern and north-eastern part of the country. Hence, continuous surveillance and ranking the hydrochemical properties are essential to evaluate the water quality status and to defend the further deterioration in study area. So the present study aims to investigate the hydrochemical characteristics of both surface and groundwater to determine the quality factors and find out the hydrogeochemical process of Sylhet, north-eastern part of Bangladesh. The study also intended to delineate the spatial distribution of susceptible parameters for future management perspective.

2 Materials and methods

2.1 Study area

The study area of Sylhet, is located in the north-eastern part of Bangladesh. Geographically it lies between 24°36′ and 25°11′ north latitudes, and 91°38′ and 92°30′ east longitudes (Fig. 1). The total area of the district is 3090.40 km^2, with the population of 2.6 million (BBS 2011). The study area is situated at Surma and Kushiyara floodplain along with the physiographic setting of the tertiary hilly regions, which represent irregular geomorphic pattern. Land use patterns are mainly depressions, agricultural lands, and settlement area in this area. Climate is one of the most important factors for the occurrence and movement of groundwater (Islam et al. 2017c). The area has a subtropical humid climate with a hot and rainy summer season and a distinct cooler dry season (Islam et al. 2017a). The temperature varies from 4 (January) to 39 °C (April) with an annual average of 25 °C. However, the north-eastern part of Bangladesh is renowned for the highest precipitation and received annual average rainfall 3000–5000 mm (Munna et al. 2015). Moreover, the relative humidity varies between 60% in the dry season and 88% in monsoon season. About 90% of extracted groundwater in the study area were used for agricultural purpose while remaining 10% is used for human consumption. Generally, groundwater from both shallow and deep aquifers is the primary sources of water for local people (Islam et al. 2017a).

2.2 Geological and hydrogeological settings

Geologically, the study area is a part of Surma basin, a sub-basin of the Bengal basin. The basin comprises a succession of thick sediments (± 16 km) which is deposited during continual marine transgression and regression events that formed huge economic deposits (Alam et al. 2003). An immense marine transgression has occurred in the Surma basin during the Holocene period surrounding a major region of the Ganges–Brahmaputra–Meghna (GBM) river delta complex with supratidal sediments (Khan et al. 2000). Lithological characteristics of the area is quite discontinuous in nature, with less extensive to broad layers having poor to moderate yielding aquifer capacity (Islam et al. 2017a). The Holocene aquifers of the study area thereby is well structured and is composed of gray fine-grained sands with peat, organic matter and gravel which is overlaid by silt and clay particles (Halim et al. 2010a). Based on subsurface geological information, it appears that

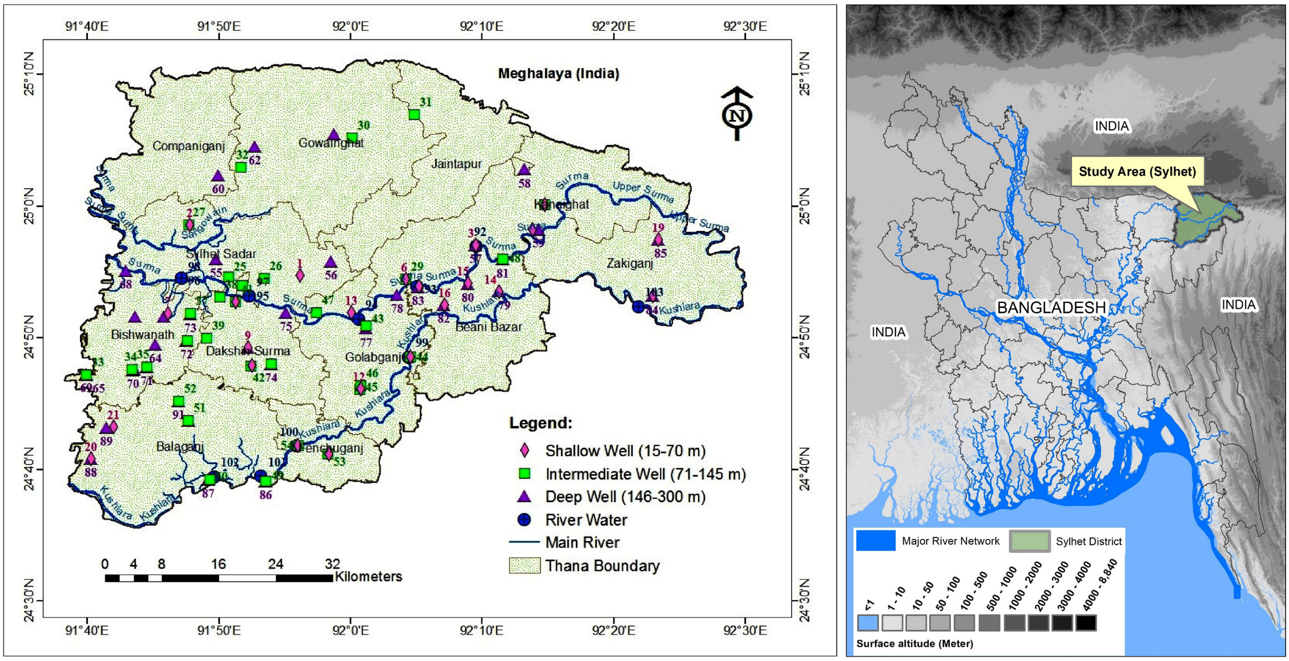

Fig. 1 Map of study area, showing the sampling locations

the thickness of a single aquifer varies from 20 to 98 m in some areas and considered as moderately good aquifer. Figure 2 shows the hydrogeological cross-section of the study area indicating the configuration of the multi-layered aquifers. Hydrogeology and aquifer layers are structurally controlled; the substantial lateral variations in the lithological characteristics of sedimentation occur to short distances which cause changes in the spatial patterns of the aquifer geometry even within 100 m depth (Islam et al. 2017a). The aquifer transmissivity in the study area varies from 146 to 825 m^2/day and coefficient storage varies from 1.5×10^{-3} to 3.48×10^{-2} (MOA 1997), inherited by

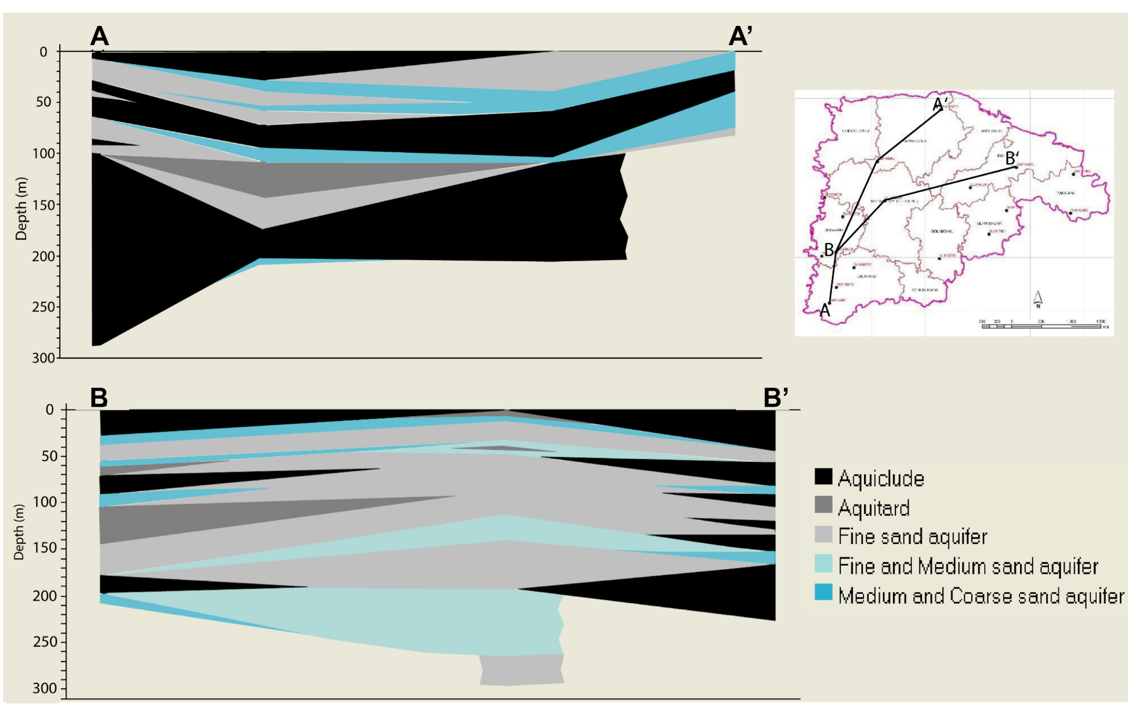

Fig. 2 Hydrogeological cross-section of study area showing the configurations of aquifers. *Source*: Islam et al. (2017a)

Holocene to late Pleistocene alluvial sands with low aquifer hydraulic conductivity (30–50 m/day) (BGS and DPHE 2001). The alluvial aquifer which is semi-confined to unconfined in nature gets endured by excessive extraction of groundwater (BGS and DPHE 2001). On the other hand, in post monsoon period the flood water and runoff from hilly regions due to excessive rainfall increase water flow in the river, stream and pond which act as a source for recharge to the aquifers (BGS and DPHE 2001). During the dry season, overexploitations accelerate the declination of groundwater level (Islam et al. 2017a).

2.3 Sample collection and analytical procedure

Field survey and sampling included reconnaissance surveys, preparing the well inventories and collection of 91 groundwater samples were done during the dry and wet seasons of the years 2014–2015 from shallow wells (depth range 15–70 m), intermediate wells (depth 71–145 m) and deep wells (depth 146–300 m) from the study area. To understand the influence of surface water, 12 samples were also collected from two major rivers in Sylhet, namely, Surma and Kushiyara. GPS readings (latitude and longitude) of the sampling points were recorded using GARMIN 12XL, GPS machine. Before sampling, the well purging was performed using the RediFlo2 submersible pump. Physicochemical properties such as pH, redox potential (Eh), electrical conductivity (EC), dissolved oxygen (DO), and temperature (°C) of water samples were measured on-site. Portable standard digital equipment of HACH brand was used to measure the physical parameters of the water samples. Field meters were calibrated using appropriate standards. Alkalinity was measured by pH-titration with H_2SO_4 using digital titrator and pH meter (USGS 1998). The samples for hydrochemical analysis were filtered through 0.45 μm membrane filters. All samples for cation analysis were acidified to pH $\approx$ 2 with HNO_3 in the field in order to avoid any precipitation of trace elements. But, the samples taken for anion analysis were not acidified. Hydro-chemical analyses were performed for major cations and anions; and other trace elements. The major cations (Na^+, K^+, Ca^{+2} and Mg^{+2}) and trace elements (Fe and Mn) of water samples were analyzed by flame atomic absorption spectrophotometer (AAS) using acetylene and air gas mixture. Arsenic (As) was measured by hydride generation method. The major anion concentrations (Cl^-, SO_4^{2-}, and NO_3^-) and DOC were determined by standard methods (APHA 1998) using an UV–VIS spectrophotometer and TOC Analyzer respectively.

2.4 Statistical analysis

Multivariate statistical techniques, i.e., correlation analysis, principal component analysis (PCA), and cluster analysis (CA) were applied to determine the solutes origin in the groundwater. For example, Pearson's correlation matrix was carried out to evaluate the similar or dissimilar origin of parameters measured in the sampled groundwater. PCA and Hierarchical CA were used to classify the sampled groundwater based on their geochemical characteristics (Meng and Maynard 2001; Hamzaoui-Azaza et al. 2011; Wu et al. 2014). PCA is one of the most popular tools to determine the geochemical weathering events, related to groundwater mineralization and differentiate the major factors like natural and anthropogenic processes influencing groundwater quality (Meng and Maynard 2001; Kim et al. 2005; Papatheodorou et al. 2007; Bouzourra et al. 2015a, b). Piper diagram was used to supplement the PCA in order to determine the major water groups in the aquifers and in the river waters (Melloul and Collin 1992; Naseem et al. 2010). Cluster analysis was further introduced to delineate groups of samples with the content of similar hydro-chemical parameters (Panda et al. 2006; Bodrud-Doza et al. 2016) and to provide supportive information to the results obtained from PCA. CA was calculated by Ward-algorithmic method and squared Euclidean distance was considered to identify the distance between clusters of similar metal contents (Bhuiyan et al. 2016). Moreover, all the maximum, minimum, mean values and standard deviation of water quality parameters obtained from the analysis were calculated and compared with Bangladesh and Indian standards.

3 Results and discussion

3.1 General hydrochemistry

Basic statistical of the physicochemical parameters and trace metal concentrations of the collected water samples along with the comparison against standards of DOE (1997), BIS (2012) and WHO (2011) are summarized in Table 1. Results show that, the mean value of the temperature, EC, DO, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^- and SO_4^{2-} were found within the range of permissible limits. However, pH indicates the strength of the water to react with the acidic or alkaline materials presents in water, which controlled by the CO_2, CO_3^{2-}, and HCO_3^- concentrations (Islam et al. 2017c). The pH values found in the shallow (3.65–7.12, mean: 5.69), intermediate (5.06–8.06, mean: 6.40), deep wells (5.32–8.38, mean: 6.64) and in surface water (5.40–8.38, mean: 6.43) vary from acidic to slightly basic in nature depicting that some pH values were

Table 1 Descriptive statistic of the measured physicochemical parameters and trace metals from the collected water samples of study area

Water samples	pH	Eh (mV)	Temp (C)	EC (μS/cm)	DO (mg/L)	DOC (mg/L)	Na^+ (mg/L)	K^+ (mg/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Fe (mg/L)	Mn (mg/L)	As (μg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	HCO_3^- (mg/L)	SiO_2^- (mg/L)
Shallow wells (SW) (n = 23)																		
Minimum	3.65	− 11.80	24.40	36.80	0.17	1.00	8.97	1.11	0.18	0.43	0.01	0.01	0.03	1.05	0.00	0.05	10.00	11.68
Maximum	7.12	178.90	27.10	730.00	5.52	42.50	89.28	4.50	23.50	17.00	18.10	0.66	148.00	132.12	25.86	5.62	287.00	66.45
Mean	5.69	64.33	25.64	292.15	1.11	10.96	39.48	2.33	7.62	4.83	8.73	0.21	39.30	21.23	5.70	1.40	123.65	34.50
SD	0.95	51.65	0.75	191.47	1.03	11.45	24.30	1.01	6.49	4.33	6.63	0.19	46.51	26.33	7.75	1.57	82.60	15.68
Variance	0.90	2667.81	0.56	36,660.94	1.06	131.07	590.71	1.02	42.12	18.76	43.98	0.04	2163.16	693.20	60.05	2.46	6823.15	245.85
Intermediate wells (IW) (n = 30)																		
Minimum	5.06	− 50.20	23.60	133.00	0.12	1.29	10.67	0.60	0.02	0.51	0.01	0.01	0.03	1.63	0.01	0.05	66.00	10.26
Maximum	8.06	99.60	27.70	641.00	6.06	20.39	127.42	13.28	34.22	14.19	22.70	0.76	115.00	18.00	55.23	16.32	312.00	78.84
Mean	6.40	29.49	25.27	287.12	1.28	4.90	47.07	2.49	8.43	4.44	6.54	0.32	25.30	8.87	2.88	2.16	157.60	35.67
SD	0.74	35.74	0.87	131.62	1.51	4.09	28.92	2.42	8.11	3.10	6.77	0.21	32.48	5.65	9.96	3.30	73.52	16.95
Variance	0.54	1277.47	0.76	17,323.04	2.27	16.76	836.33	5.86	65.76	9.61	45.87	0.05	1055.07	31.94	99.28	10.87	5405.49	287.27
Deep wells (DW) (n = 38)																		
Minimum	5.32	− 83.60	23.00	109.00	0.11	1.65	5.51	0.64	0.08	0.75	0.01	0.02	0.12	0.83	0.00	0.08	52.00	10.26
Maximum	8.42	89.90	28.70	726.00	4.93	34.91	184.84	8.14	22.19	8.65	15.95	0.83	72.50	60.00	19.10	27.12	262.00	57.83
Mean	6.64	15.90	25.53	302.47	0.79	5.02	51.90	2.36	5.93	3.52	5.91	0.30	21.38	9.68	1.91	4.51	154.34	33.42
SD	0.75	41.34	1.32	128.52	0.82	5.49	35.57	1.59	4.32	1.91	4.86	0.23	19.17	9.96	3.34	7.19	62.09	13.19
Variance	0.57	1709.13	1.75	16,516.42	0.68	30.16	1265.43	2.52	18.64	3.66	23.63	0.05	367.30	99.18	11.15	51.64	3854.99	173.87
River water (RW) (n = 12)																		
Minimum	5.40	− 60.10	21.10	50.10	5.26	2.26	2.95	0.94	2.67	1.75	0.01	0.001	–	3.06	0.20	0.34	19.00	10.39
Maximum	8.38	73.00	28.70	200.00	7.29	6.29	9.70	4.98	20.75	5.54	0.42	0.10	–	16.11	5.67	29.13	60.00	74.40
Mean	6.43	22.78	26.12	91.55	6.03	4.17	5.57	1.78	6.42	3.39	0.10	0.03	–	8.69	1.14	6.55	33.83	27.68
SD	0.88	46.40	2.59	42.84	0.69	1.48	2.02	1.46	4.88	1.28	0.12	0.03	–	3.19	1.86	8.96	13.31	21.61
Variance	0.77	2152.80	6.73	1835.15	0.48	2.20	4.09	2.14	23.81	1.64	0.02	0.001	–	10.17	3.46	80.32	177.24	467.14
Standards																		
Bangladesh standard: DOE (1997)	6.5–8.5	–	20–30	1000	6	–	200	12	75	30–35	0.3–1.0	0.1	50	150–600	10	400	–	–
Indian standard: BIS (2012)	6.5–8.5	–	–	–	–	–	–	–	75	30	0.3	0.1–0.3	10–50	250–1000	45	200–400	–	–
WHO (2011)	6.5–8.5	–	–	1500	–	–	200	12	75	30	0.3	0.1	10	250	45	250	–	–

found to be within the permissible limit and some are below of permissible limit indicates acidic water (Table 1). More specifically, lower pH values (< 5) were found in some shallow wells i.e., SW_1, SW_13, SW_17 and SW_22 provided in Table S1 of supplementary information. Acidic water may significantly change the groundwater chemistry and dissolved solutes more actively (Premalal and Jayewardene 2015), which may evolve from the biogeochemical process, dissociation of humic acids, oxidation of sulphur and nitrogen compounds and anthropogenic sources (Knutsson 1994). Study also found that, shallow wells groundwater is characterized by the low HCO_3^- concentration, which may be as a result of the runoff waterways, that continue to moisturize the area and eventually leading to a decrease in pH (Nelson 2002). Electrical conductivity (EC) of groundwater depends upon temperature, ionic concentration and types of ions present in the water (Kumar et al. 2014; Islam et al. 2017c). The EC concentrations (μS/cm) in shallow (37–730, mean: 292), intermediate (133–641, mean: 287), deep wells (110–726, mean: 302) and in surface water (50–200, mean: 92) were found to be low, indicating the presence of low contents of dissolved ions in the surface and groundwater of the study areas. Moreover, dissolved oxygen (DO) in surface water was found to be in good range (5.26–7.29 mg/L, mean: 6.03 mg/L), which indicates no significant pollution load of surface water in study areas. Again in shallow, intermediate and deep wells have DO level in the range of 0.17–5.52 mg/L, 0.12–6.06 mg/L, 0.11–4.93 mg/L respectively, indicates anoxic–oxic in condition. Furthermore, the concentration of dissolved organic carbon (DOC) in the collected groundwater samples were recorded (1.0–42.5 mg/L, mean: 10.96 mg/L) for shallow wells (1.29–20.39 mg/L, mean: 4.90 mg/L) for intermediate wells (1.65–34.91 mg/L, mean: 5.02 mg/L) for deep wells and (2.26–6.29 mg/L, mean: 4.17 mg/L) for surface water. Organic matter presence in the sediments might have played a significant role to generate anaerobic environments in the aquifers (Ahmed et al. 2004; Halim et al. 2010b).

In shallow, intermediate and deep wells the variation of mean cation concentration (mg/L) were found in same order by $Na^+ > Ca^{2+} > Mg^{2+} > K^+$, while in surface water the order is $Ca^{2+} > Na^+ > Mg^{2+} > K^+$. Again, the variations of the mean anion concentrations (mg/L) in shallow and intermediate wells were found in order of abundance as; $HCO_3^- > Cl^- > NO_3^- > SO_4^{2-}$ while the mean anion concentrations (mg/L) in deep wells and surface water reveal order of abundance as; $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$. Elevated presence of SiO_2 (mg/L) in shallow (11.68–66.45, mean: 34.50), intermediate (10.26–78.84, mean: 35.67) and deep wells water (10.26–57.83, mean: 33.42) as well as in surface water (10.39–74.40, mean: 27.68) may be attributed to the influence of silicate weathering in the groundwater and river water of the study area (Halim et al. 2010a).

Piper diagram (Fig. 3) was used to present and classify the major ions of groundwater and surface water, and to summarize the main contrasts in hydrochemical composition between different water sources. Interpretation of the hydrochemical data implies that, in most cases shallow, intermediate and deep groundwater is Na–HCO_3 type. Water samples plotted at the lower corner of the diamond is composed primarily of Na^+, K^+ and HCO_3^-. The plot according to this arrangement is presented in Fig. 2, where three classes of combinations were obtained in groundwater. The position of the groundwater in the anions triangle indicates dominance of HCO_3^-, but the absence of SO_4^{2-} in the groundwater. HCO_3^- is the dominant anion and its concentration is mainly attributed to weathering as well as organic matter degradation. Na^+ and HCO_3^- concentrations comes mainly from the weathering of alkali-feldspars related with the recharge areas (Sakram and Adimalla 2018). However, Na–HCO_3 represents 'Fresh Type' water in the aquifer. The other two types of Blended Water are found in the groundwater of Sylhet City Corporation and Gowainghat areas. The three intermediate tube wells of mid-Holocene aquifer in Sylhet City Corporation area have shown the water type of Ca–Mg–Na–HCO_3, which is low mineralized water indicating the initial source of water recharging into the aquifer systems. Meanwhile, Na–Ca–Mg–HCO_3 blended type groundwater observed in Pleistocene aquifer well at Gowainghat, showed slightly increase of Na^+ concentration with respect to Ca^{2+} and Mg^{2+} concentrations. The increase in Na^+ exchange for Mg^{2+} suggests softening process, which may

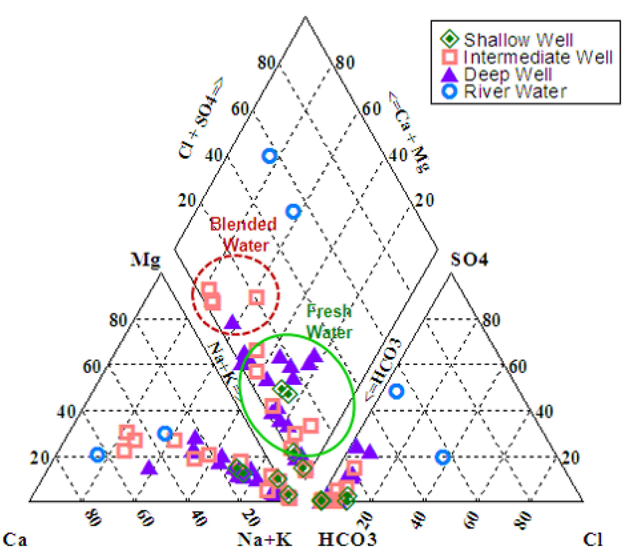

Fig. 3 Piper Trilinear diagram for the surface and groundwater samples of study area

indicate much more water–rock interactions along the flow paths (Bhuiyan et al. 2015). The water types for Kushiyara and Surma rivers are of Ca–Mg–SO_4–HCO_3 and Ca–Mg–HCO_3 type indicating the dominance of Ca^{2+} and Mg^{2+} in the cations and interplay of SO_4^{2-} and HCO_3^- anions which influence the water quality in the study area.

3.2 Hydrogeochemical processes

Geochemical processes like rock–water interaction, and evaporation is the wide classification of lithogenic influences e.g., silicate weathering, carbonate dissolution, and evaporate dissolution (Kumar 2014). The results of these lithogenic influences can be obtained by sketching the bivariate plots of Ca^{2+}/Na^+ versus HCO_3^-/Na^+ and Ca^{2+}/Na^+ versus Mg^{2+}/Na^+ (Fig. 4a, b). Both of these plots illustrates that, carbonate dissolution is almost absent in the groundwater aquifers. The major influential process is found to besilicate weathering as well as evaporative dissolution that controls the hydrochemical solute contents of the groundwater in study area (Kumar 2014). Further bivariate plots of ($Ca^{2+} + Mg^{2+}$) versus HCO_3^- and ($Na^+ + K^+$) versus HCO_3^- were introduced to comply with the influence of silicate weathering in the groundwater of the study area (Fig. 5a, b), whereas silicate weathering in groundwater could be generated from alumino-silicates (Lasaga 1984; Wanda et al. 2011).

Cation exchange process in the groundwater of the study area was also investigated by introducing a bivariate plot between Cl^- correlated ($Na^+ + K^+$) and ($Ca^{2+} + Mg^{2+}$) correlated ($HCO_3^- + SO_4^{2-}$) (Fig. 6). If functioning cation exchange within Na^+ and ($Ca^{2+} + Mg^{2+}$) is active in an aquifer, the slope which is drawn from the plot would be -1 (i.e., $y = -x$). The slope plot found for the study area (-0.77) exhibits that, cation exchange is a predominating process in many samples (Kumar 2014).

Elemental relationship was assessed to determine the major elements contributing to the water mineralization (Fig. 7a–d) Ca^{2+} versus SO_4^{2-} plot (Fig. 7a) and ($Ca^{2+} + Mg^{2+}$) versus ($SO_4^{2-} + HCO_3^-$) cation exchange plot (Fig. 7b) indicates the gypsum dissolution influenced by irrigational return flow or by saturation or oversaturation of calcite and aragonite bearing minerals (Kraiem et al. 2013). The bivariate plot of Na^+/Cl^- generally is used to determine the mechanism of salinity, rock–water interaction and saline intrusions (Sivasubramanian et al. 2013). Most of the collected samples lay close to line (Fig. 7c) indicating that NaCl exhibits a continuum from precipitation and uncontaminated groundwater (Panno et al. 1999). However, a few samples possess little Cl^-, which indicates that the enrichment of Na^+ is due to rock–water interaction within the aquifer (Panno et al. 1994; Hackley 2002) or, contamination of animal and human waste in the study areas (Panno et al. 1999). Bivariate plot of Na^+/HCO_3^- (Fig. 7d) indicated the increased concentration of Na^+ compared to HCO_3^-. This relation suggests that, aquifer sediments in the study area is influenced by carbonate weathering (Bhuiyan et al. 2015).

3.3 Trace elements in water

Concentrations of trace elements like Fe, Mn and As were found to be higher in the aquifers of study area. Table 1 shows that the mean values of Fe in groundwater for shallow (8.73 mg/L), intermediate (6.54 mg/L) and deep wells (5.91 mg/L) exceed DOE of Bangladesh standard (1997) BIS of Indian standard (2012) and WHO (2011) standards. The spatial distribution (Figs. 8 and 9) of Fe is plotted for the shallow, intermediate and deep wells in

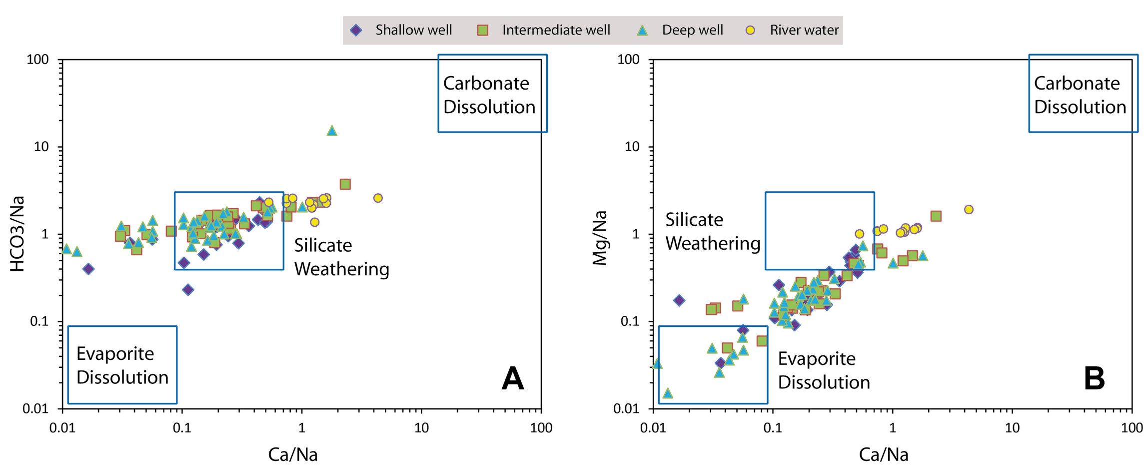

Fig. 4 Bivariate plot **a** Ca^{2+}/Na^+ versus HCO_3^-/Na^+ and **b** Ca^{2+}/Na^+ versus Mg^{2+}/Na^+ to identify the minerals weathering of water in study area. The boxes represent the ranges of approximate compositions of the three main source end members (evaporate dissolution, silicate weathering and carbonate dissolution) without any mixing

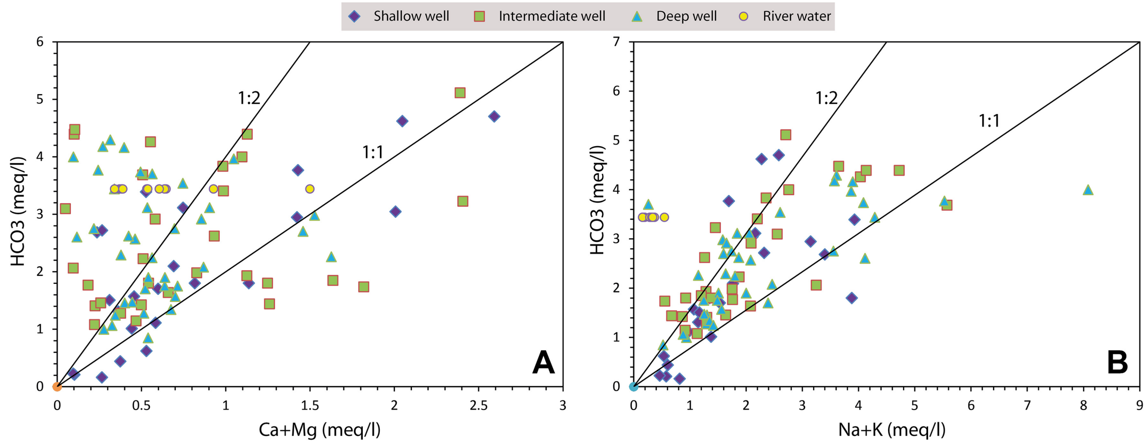

Fig. 5 Bivariate plots of **a** $Ca^{2+} + Mg^{2+}$ versus HCO_3^- and **b** $Na^+ + K^+$ versus HCO_3^- to identify the silicate weathering in study area

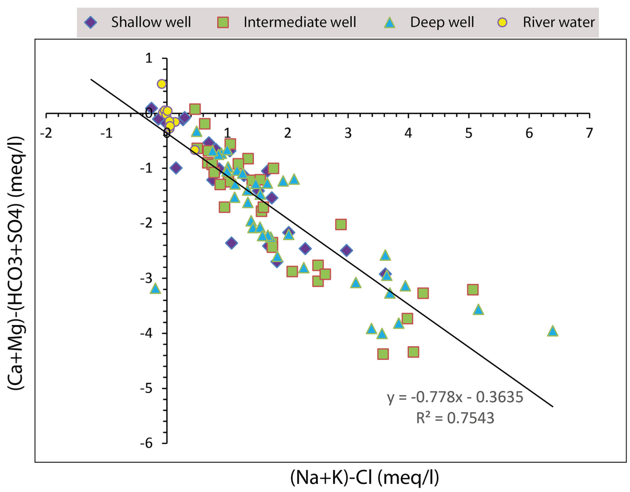

Fig. 6 Bivariate plot of Cl^- corrected $Na^+ + K^+$ and $Ca^{2+} + Mg^{2+}$ corrected $HCO_3^- + SO_4^{2-}$ to identify the cation exchange of water in study area

order to determine the variability of Fe in different type of aquifers; and it was found that iron is very high in deep wells compared to the shallow and intermediate wells. The range of Fe in deep wells are found 0.01–15.95 mg/L, and only 24% samples of deep wells falls within the permissible limits of Bangladesh standard DOE (1997). Iron is dominantly high in both intermediate and deep wells of Gowainghat Upazila located at the northern part of the study area, ranging from 10.3 to 21.3 mg/L. In between the intermediate depth, the highest value of Fe is found in the sample IW31 (21.3 mg/L) (Table S1). In most of the cases, iron is dominant trace element in the deep aquifer and these high iron values might have created aesthetic and others health problems to the inhabitants. The consumption of excessive Fe could lead to 'chromatosis', a severe disease that can destroy the different body organs (WHO 2003). Other symptoms i.e., fatigue, loss of weight, and joint pain also could occur due to excessive presence of Fe in drinking water (Karen 2017).

However, the mean values of Mn in groundwater for shallow (0.21 mg/L), intermediate (0.32 mg/L) and deep wells (0.30 mg/L) cross the all standard limits (Table 1). This high concentration of Mn can cause weakness, anorexia, muscle pain, and apathy among the native people of the study area (WHO 2011). Manganese is also unlikely responsible for cancer or reproductive damage (USEPA 2004). The spatial distribution of Mn is plotted for all the wells to see the variability of Mn in the shallow, intermediate and deep aquifers throughout the study area (Fig. 10). Almost 80% intermediate and 68% deep wells, exceed the Bangladesh standard limit 0.1 mg/L, having highest value in the north-western part of study area. The samples of the Kushiyara and Surma Rivers contain low level of Mn, due to presence of oxidizing environment in river water. The variation of Mn concentrations at different depth is presented in Fig. 11. Relatively high concentrations of Mn was observed in some deep groundwater samples in the area [sample id: DW_62 (0.83 mg/L), DW_63 (0.74 mg/L), sample id: DW_73 (0.71 mg/L)] and the plotted Mn distribution indicates that the deep aquifer is gaining risk for manganese in the drinking water.

Furthermore, the groundwater of the study area is found to be contaminated by arsenic (As), and crossed the permissible limits of all standards. The mean concentrations of As in shallow, intermediate and deep wells were found 39.3, 25.3 and 21.4 μg/L respectively, and varied from 0.03 to 148 μg/L (Table 1). The highest concentrations of As in shallow, intermediate and deep wells were found in the SW_21 (148.0 μg/L), IW_50 (89.1 μg/L), DW_88 (72.5 μg/L) (Table S1). However, the elevated level of As concentrations can cause severe potential human health risks like vomiting, abdominal pain, skin cancer, muscle cramping, cancer in bladder and lungs in the residents of the study area (WHO 2017). The spatial distribution of As

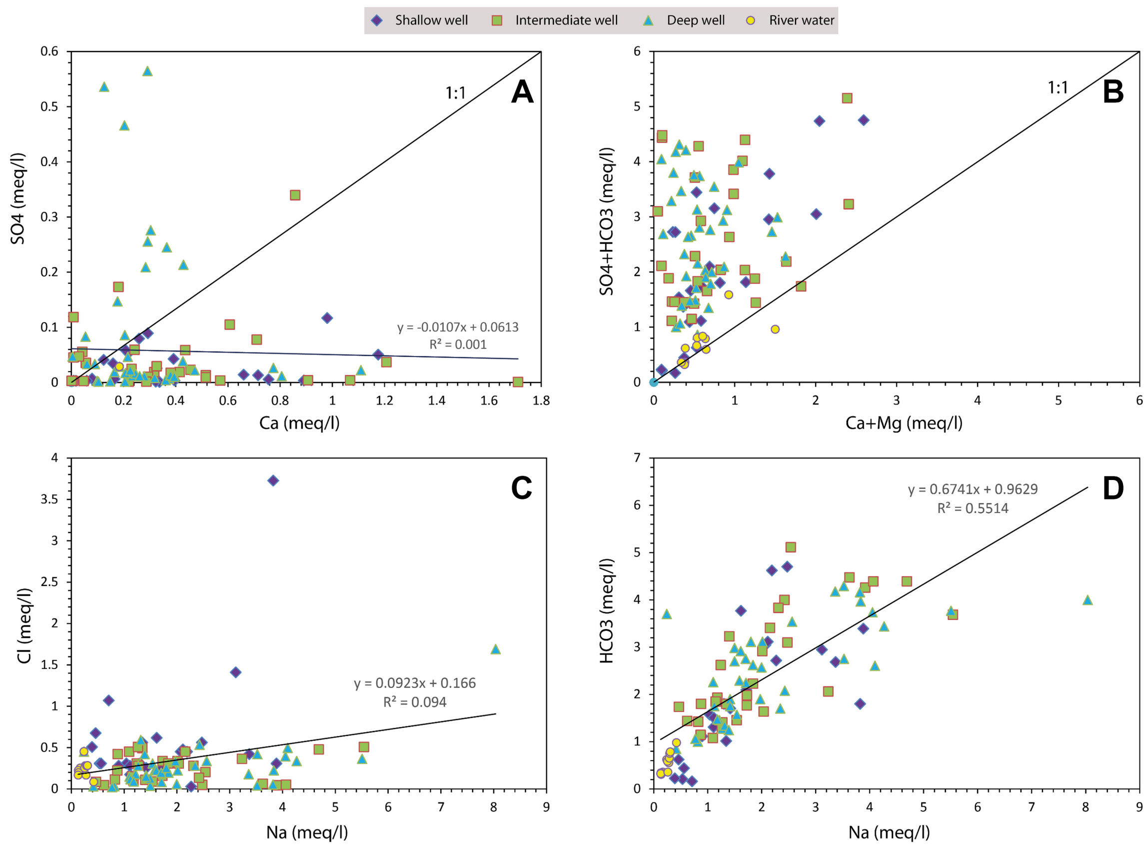

Fig. 7 Bivariate plots showing the relationships between **a** Ca^{2+} and SO_4^{2-}, **b** $Ca^{2+} + Mg^{2+}$ and $SO_4^{2-} + HCO_3^-$ **c** Na^+ and Cl^- **d** Na^+ and HCO_3^-

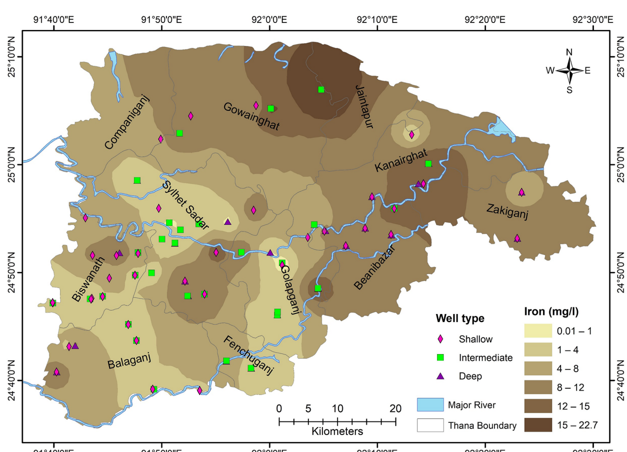

Fig. 8 Spatial distribution of Fe concentration of groundwater in Sylhet area

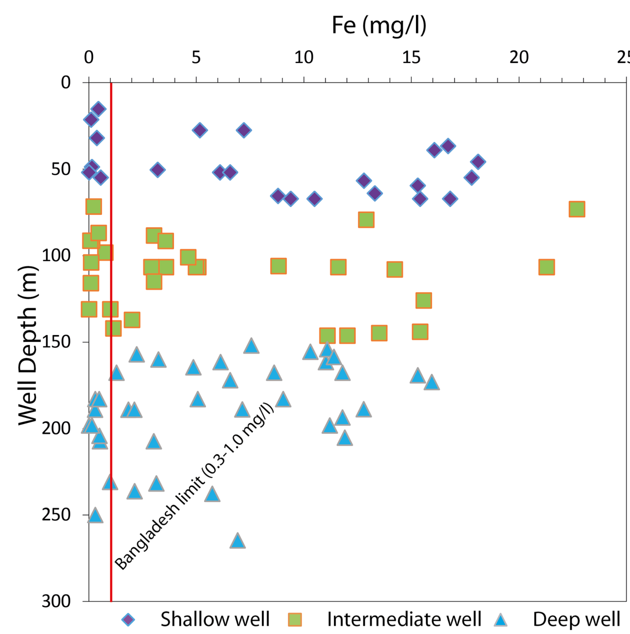

Fig. 9 Variation of Fe at different depths of aquifers in Sylhet area

in the groundwater of Sylhet has been plotted in Fig. 12, and it is found that, south-western and northern part of the study area is highly contaminated. It is also noted that, shallow and intermediate aquifers are highly contaminated and most of the deep wells are free from As contamination (Table S1). Several hypotheses suggest that, As contamination in groundwater aquifers of Bangladesh might be attributed due to the microbial and chemical reductive dissolution of As-bearing iron oxides in the aquifer

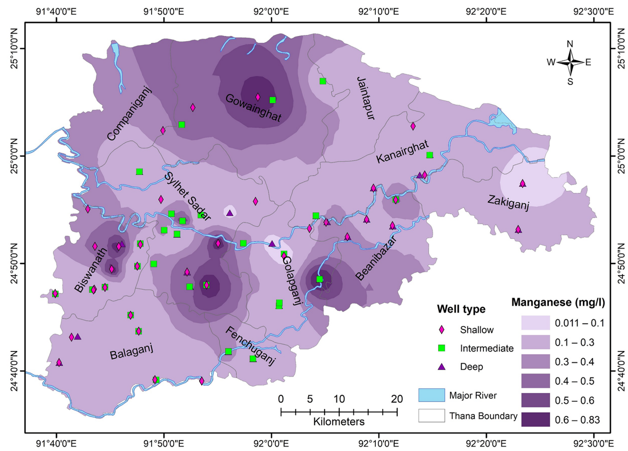

Fig. 10 Spatial distribution of Mn concentration of groundwater in Sylhet area

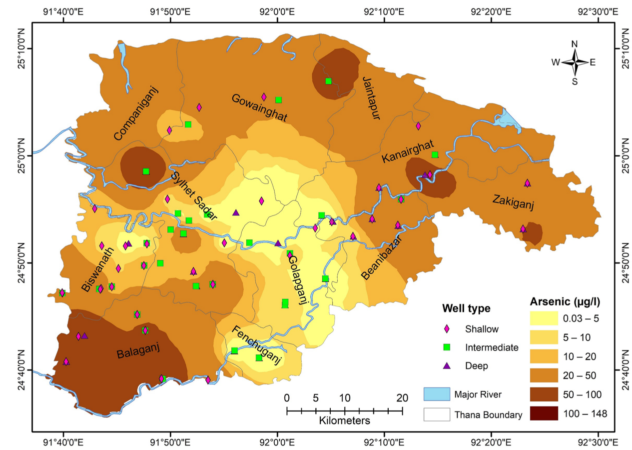

Fig. 12 Spatial distribution of As concentration of groundwater in Sylhet area

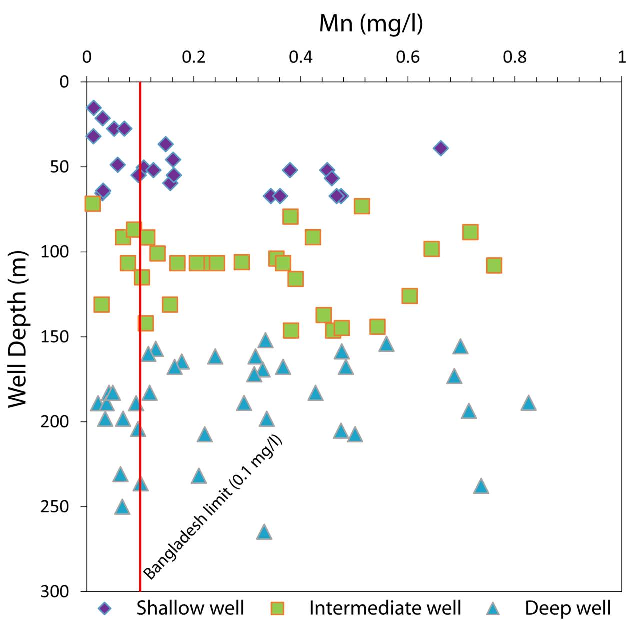

Fig. 11 Variation of Mn at different depths of aquifers in Sylhet area

sediments (Nickson et al. 2000; Islam et al. 2004; Ravenscroft et al. 2005; Stollenwerk et al. 2007). Occurrences of organic matter in the Bengal basin aquifer sediments had also been identified by many researchers (Nickson et al. 2000; McArthur et al. 2001; Ahmed et al. 2004; Islam et al. 2004; Zheng et al. 2004). This organic matter depletion could accelerate the redox reaction orders in the aquifers that enhance the mobilization of As and a high amount of DOC facies may sustain for a longer period in the groundwater phase (Halim et al. 2010b). Some recent studies regarding Bangladesh perspective have revealed moderate to strong correlation of DOC with Fe and As which is believed to be resulted from microbial mediated reductive dissolution of FeOOH along with adsorbed As (Halim et al. 2009; Ahmed et al. 2004). Lower DO levels in the aquifers also might be the action of organic matter presence in the sediment, which plays a significant role to generate anaerobic environments in the aquifer (Halim et al. 2010b).

The bivariate plots for As correlation with DOC, Fe and Mn are illustrated in Fig. 13a–d. No significant correlation was shown among the plots As versus DOC, Fe versus DOC, As versus Fe and As versus Mn. This lack of correlation delineated that, As mobilization especially in the shallow and intermediate aquifers might be affected by the multiple mechanisms like microbial activities or chemical reductive dissolution of As-bearing FeOOH in aquifer sediments (Halim et al. 2010b). The aquifers in this case are in mid-Holocene stage, where the aquitard or clay lenses are absent. These cases do not necessarily imply the contamination of intermediate aquifers. Major element compositions of groundwater often depend on hydrologic factors (e.g. residence time, evaporation, mixing with adjacent aquifers) as well as geochemical processes, namely mineral dissolution and ion exchange (Aggarwal et al. 2000). The vertical distributions of As contents at different depths of groundwater samples are presented in Fig. 14. It was found that, As concentration of groundwater decrease with the increasing depth. However, there are 7 intermediate and 3 deep tube wells were also found to have As concentration higher than the Bangladesh standard limit 50 µg/L. These high concentrations might have occurred due to construction fault of wells or/and greater permeability of the shallow aquifers (Halim et al. 2010b). As the concentration of trace elements were found very negligible in surface water, with mean value of Fe (0.1 mg/L), Mn (0.03 mg/L) and As (BDL) it is assumed that, there is no influence of surface water to contaminate the aquifers.

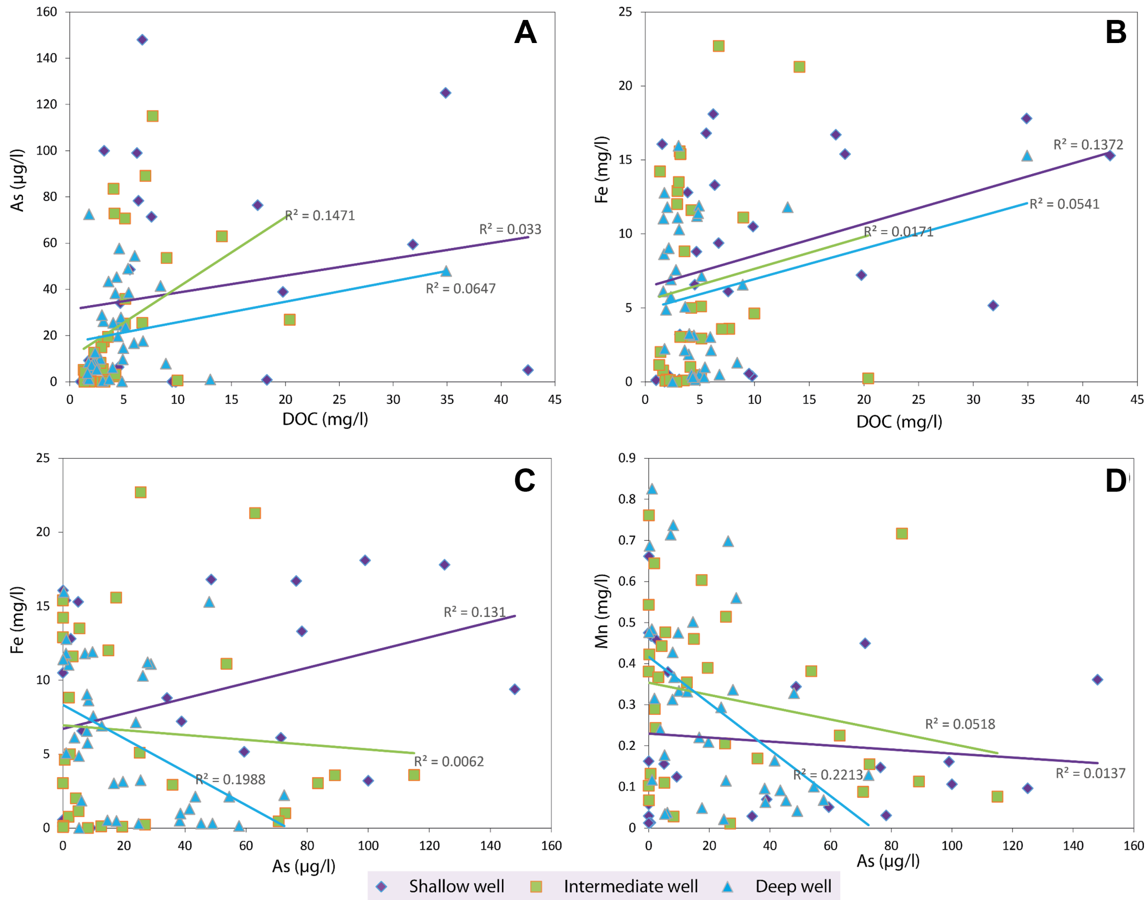

Fig. 13 Bivariate plots showing the relationships between **a** As and DOC **b** Fe and DOC **c** Fe and As and **d** Mn and As at different depth

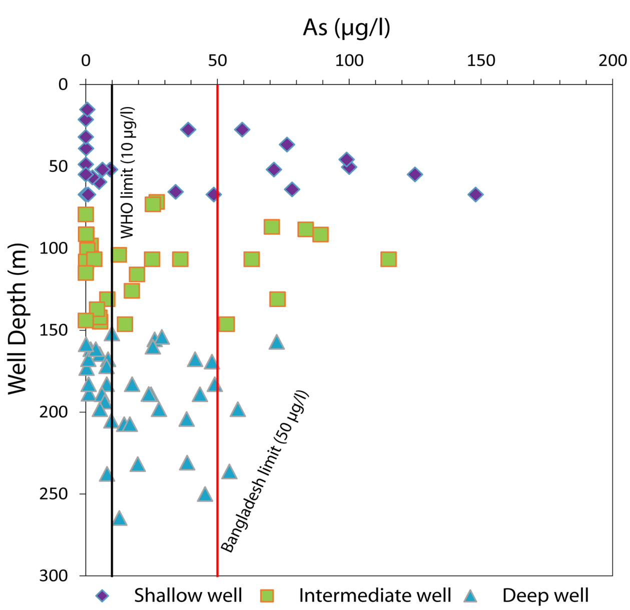

Fig. 14 Variation of As at different depths of aquifers in Sylhet area

3.4 Results of statistical analysis: factors influencing water chemistry

Pearson's correlation matrix was used to determine the interrelationships among different water quality parameters in both groundwater and river water (Tables S2 and S3). For groundwater, strong positive and negative correlations were found for Na–EC (0.753), HCO_3–EC (0.882), Mg–Ca (0.837) and Eh–pH (– 0.957). Moderate positive and negative correlation were found in HCO_3–pH (0.626), EC–Eh (– 0.5), Na–Eh (– 0.555), HCO_3–Eh (– 0.636), As–EC (0.617), HCO_3–Na (0.681), NO_3–Mg (0.563), Mn–Fe (0.506), SiO_2–Fe (0.573), HCO_3–As (0.541). Positive correlations indicate the same origin that could be coming from natural environment or from human activities due to source mobilization (Haloi and Sarma 2012). For instance, strong positive correlations of EC–Na (0.753) indicate high presence of dissolved ions in the study area. In addition, moderate significant correlation for example As–HCO_3 (0.541) in groundwater indicates the mobilization of As in groundwater within the anoxic environment (Halim et al. 2010b). Again moderate significant positive correlations (e.g. Fe–Mn: 0.506) indicate the origination from the same points. Similar findings were also found in shallow groundwater of Rangpur district (Islam et al. 2017b) and Sylhet region (Islam et al. 2017a); where redox levels are controlled by trace elemental occurrence and the characteristics of rocks beneath the aquifer (Islam et al. 2017a). Moreover, the insignificant negative correlation (e.g. HCO_3–Mn: − 0.287) explain that, these parameters were independent from each other in origin (Kamrani et al. 2016). Similar kind of results was also revealed for river water (Table S2). Results like significant correlations between K–SO_4 (0.838) and K–NO_3 (0.543) indicates the influence of excessive agricultural practices with high precipitation along and heavy discharge from upstream occurred in the study area (Shammi et al. 2017).

Varimax Principal Component Analysis (PCA) for both R-mode and Q-mode were further applied to identify the contributing factors of measured parameters influencing the groundwater (Table 2) and (Table S3) of supplementary material. Total six factors of R-mode with Eigen values > 1 were extracted for groundwater data set that displayed 76.16% of total variance (Table 2). The scree plot was also introduced to understand the principle component numbers in the underlying parameters (Fig. 15a). The positive and negative values in PCA explained that, the water samples were essentially affected or unaffected by the presence of extracted loads on a specific component. About 49.34% of total variance is displayed in the first three loadings (Fig. 15b). In this study, PCA, PC2, PC3, PC4, PC5 and PC6 explained the 20.46%, 16.81%, 12.06%, 10.55%, 8.92% and 7.33% of variance respectively. The results of R-mode analysis were also correlated with the Q-mode analysis (Table 2). The first PC (PC1) explaining 20.46% of total variance is positively loaded with pH, EC, Na^+ and HCO_3^- (Table 2). Very high loading of these elements were found in the samples of IW_39, IW_43, IW_50, DW_64, DW_65, DW_68, DW_76, DW_84, DW_88, DW_89 and DW_91 (Table S3). The geochemistry in the shallow to intermediate groundwater aquifers of these areas are influenced by silicate rock weathering (Islam et al. 2017a). In PC2 group, Ca^{2+}, Mg^{2+} and NO_3^- factors contributed most in the sampling points like SW_4, SW_5, SW_10, SW_14, SW_15, SW_18, IW_25, IW_26, IW_31, DW_56, DW_64 and in RW_92. Both Ca^{2+} and Mg^{2+} factors describe natural ion origin coming from the calcite dissolution in these areas. Again carbonate rock beneath the Holocene alluvial aquifer resulting the higher concentration of Ca^{2+} and Mg^{2+} in the groundwater of these study areas (Islam et al. 2017a). Anthropogenic factors such as agricultural practice by the native people involve excessive use of fertilizers and pesticides which increase the concentrations of NO_3^- in the shallow groundwater through percolation (Amiri et al. 2014). However, Fe and Mn mostly contributed in PC3and heavily extending in the sampling area of SW_9, SW_11, SW_16, IW_30, IW_37, IW_40, IW_42, IW_44, IW_47, IW_48, IW_53, DW_61, DW_62, DW_63, DW_73, DW_74, and

Table 2 Varimax rotated principal component analysis (R-mode) for collected water samples

Parameters	PC1	PC2	PC3	PC4	PC5	PC6
pH	0.814	− 0.088	− 0.17	− 0.295	0.312	− 0.125
Eh	− 0.824	0.074	0.206	0.228	− 0.338	0.061
Temperature	− 0.158	− 0.098	− 0.206	− 0.006	− 0.67	0.271
EC	0.781	0.374	0.072	0.405	− 0.141	0.051
DO	− 0.328	− 0.016	− 0.672	− 0.257	0.264	0.241
DOC	0.068	0.545	0.008	0.296	− 0.171	0.301
Na	0.792	− 0.102	0.056	0.449	− 0.159	0.058
K	0.058	0.261	0.217	− 0.103	0.189	− 0.737
Ca	− 0.01	0.819	0.075	− 0.11	0.138	− 0.089
Mg	0.082	0.902	0.107	0.047	0.098	− 0.112
Fe	− 0.174	0.387	0.761	0.112	0.073	0.225
Mn	− 0.079	− 0.046	0.818	− 0.201	0.215	− 0.116
As	0.404	0.363	0.029	0.629	− 0.078	− 0.13
Cl	− 0.047	− 0.007	− 0.033	0.856	0.055	0.097
NO_3	0.083	0.766	0.06	0.108	− 0.095	− 0.039
SO_4	− 0.057	− 0.034	− 0.008	− 0.005	0.789	0.075
HCO_3	0.858	0.316	0.13	0.144	− 0.175	− 0.126
SiO_2	− 0.105	0.182	0.514	− 0.066	0.127	0.63
Eigenvalues	3.683	3.026	2.172	1.9	1.607	1.321
% of variance	20.464	16.812	12.068	10.557	8.926	7.339
Cumulative %	20.464	37.276	49.344	59.901	68.827	76.166

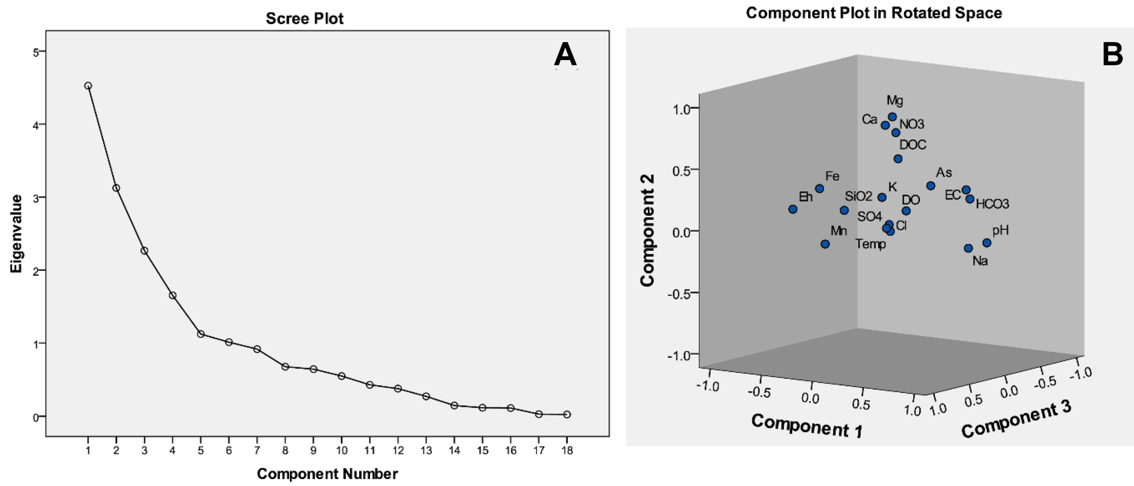

Fig. 15 Principal component analysis by **a** scree plot of the characteristic roots (eigenvalues), and **b** component plot in rotated space

in DW_78. The study revealed that, these areas might be attributed dueto the natural activities like chemical weathering of parental rock with ionic exchange (Omo-Irabor et al. 2008). The fourth PC (PC4) is mostly loaded with As and Cl^{-}, which occurred probably due to groundwater–rock interaction (Harvey et al. 2002). This kind of interaction is highly predominant in the sampling areas of SW_2, SW_5, SW_6, SW_8, SW_13, SW_20, SW_21, IW_33, DW_57, and DW_84. SO_4^{2-} is mostly occupied in PC5 and heavily extending in the SW_21, IW_32, DW_57, DW_59, DW_62, DW_66, DW_67, DW_73, RW_92, RW_97, RW_98, RW_99 and RW_102. Gypsum dissolution and high agricultural activities could be responsible factors in these areas (Islam et al. 2017a). The major occurrences in PC6 is SiO_2, which suggests that the geogenic factors like silicate weathering influence the sampling points of the study areas i.e., SW_4, SW_9, SW_16, SW_19, IW_47, IW_48, DW_57, DW_58, DW_75, DW_78, DW_84, RW_100, RW_101, and RW_102 (Bhuiyan et al. 2016; Halim et al. 2010a).

The hierarchical cluster analysis (CA) was performed, which is very similar to the PCA's results. Parameters belonging to the same cluster are likely to have originated from a same source (Bhuiyan et al. 2016). The R-mode CA indicates four clusters for analyzed parameters, presented in Fig. 16. Cluster-1 consists of EC, HCO_3, Na, As, pH indicated the influence of silicate weathering and rock–water interactions (Islam et al. 2017a). Cluster-2 includes Ca, Mg, NO_3, DOC and K indicating the influence of both natural origins like calcite dissolution and anthropogenic origin such as leaching from agricultural practices due to excessive use of fertilizers and pesticides in the study area (Islam et al. 2017a, Amiri et al. 2014). Fe, Mn and SiO_2 constitute cluster-3 indicates the chemical weathering of the parent rock in the study area (Omo-Irabor et al. 2008;

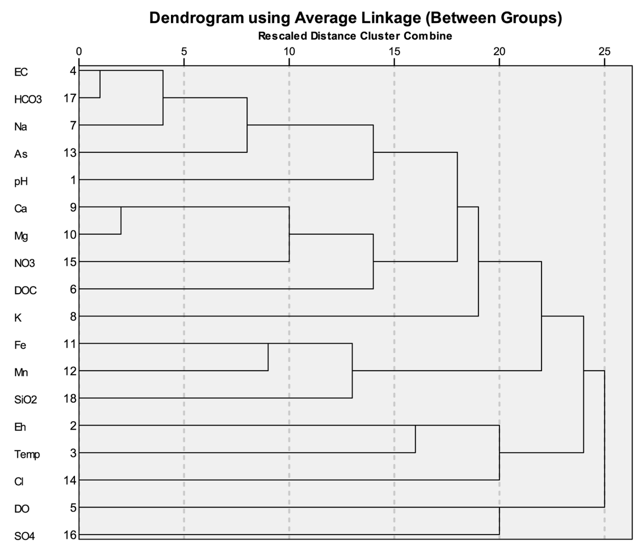

Fig. 16 Dendrogram showing the hierarchical clusters (R-mode) of analysed parameters

Islam et al. 2013). Cluster-4 constituted with Eh, temperature, Cl, DO and SO_4^{2-} is mostly related with the anthropogenic activities like acceleration of fertilizer leaching to the groundwater (Singh et al. 2011) or natural activities like evaporate mineral dissolution (Kraiem et al. 2013).

4 Conclusions

Understanding of hydrochemistry of groundwater is essential for the management of groundwater resources. In this study, statistical analysis, graphical plots, and spatial distribution of parameters were applied to investigate the hydrogeochemical characteristics and major factors influencing groundwater. Among the 18 parameters from 103 water samples it was found that Fe, Mn and As were predominant in the groundwater, creating a potential threat to human health. Most elevated levels of As were found in the shallow wells whereas the concentrations of Fe and Mn are high in a few intermediate and deep wells. Most of the groundwater are Na–HCO_3 type, and main geochemical processes included silicate rock weathering, calcite dissolution, parental rock weathering, ion exchange influenced the water chemistry. This study contributes necessary contextual information on physiochemical parameters, possible sources and controlling factors of groundwater quality and its spatial variation in the study area. Hence the results may consider for future planning, while using the groundwater for drinking purposes. However, further study is recommended to provide a more scientific basis in developing the representative strategies by characterizing the water quality according to the hydrochemical results.

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Compliance with ethical standards

Conflict of interest The authors declared that they have no conflict of interest.

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