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Research Article

Mechanism of Rock-Water Interaction in Kuzhithuraiyar Sub-Basin, Kanniyakumari District, Tamilnadu, India

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Abstract

The chemical composition of groundwater is changed by a number of hydro-geochemical processes. Rock-water interaction is the primary process that controls the ionic concentration of water, even though vaporization, concentration and dilution due to precipitation alter the chemical composition of groundwater. The chemical reactions differ depending on the preliminary composition of water, the geological formations it passes through and even how long it has been there. The 48 groundwater samples used in this study were collected, and the pH, Electrical Conductivity, Total Dissolved Solids, major cations and anions concentrations were measured. Gibb's Plot, Bivariate diagrams, Piper's Trilinear diagrams along with Saturation Indices were used to examine the mechanism of rock-water interaction in Kuzhithuraiyar Sub-Basin.

Keywords: Rockwater Interaction, Bivariate Diagrams, Gibbs Plot, Pipers Trilinear Diagram, Saturation Indices

Introduction

The chemistry of natural water is influenced by rock-water interaction. (Thakur *et al.*, 2013; Mehmandosti and Adabi, 2013; Belkhiri *et al.*, 2012; Loni *et al.*, 2015). When primary and secondary minerals are weathered, silica and cations are released into the groundwater (Adithya, *et al.*, 2016). Hydrogeochemical processes such as precipitation, dissolution, recharge, discharge, oxidation- reduction, ion-exchange, water mixing, residence time, *etc.* all affect the composition of groundwater (Reghunath *et al.*, 2002; Mallick *et al.*, 2018). The aquifer system may undergo the following changes as a result of chemical processes such as ionic concentration could go up or down, mobility of the dissolved ions might be affected and even chemical reactions could also affect the water's pH (Dehnavi *et al.*, 2011; Elango *et al.*, 2003; Deutsch, 1997; Jalali, 2005, 2006, 2007; Jalali and Khanlari, 2008). The chemical components that make up water have a big impact on how it is used for industrial, agricultural, and residential purposes (Krishnakumar *et al.*, 2017). Surface and ground water degradation is typically accelerated by both natural and human-caused factors. These include population expansion, weathering, droughts, industrial processes, soil erosion, mineral dissolution, precipitation, and agricultural practices (Kumar and Sangeetha, 2020; Misaghi, *et al.*, 2017; Gu, 2019; Sudhakaran, *et al.*, 2020).

The hydrogeochemistry of the groundwater is mostly determined by the geology and aquifer characteristics of the research area (Datta and Tyagi, 1996; Mallick *et al.*, 2018; Ahmad *et al.*, 2023). The variety in aquifer types and quantification of mineralogical composition can be used to understand various hydrochemical features (Ahmad *et al.*, 2023; Sun, 2007; Sun and Gui, 2015). In recent times, multivariate statistical analysis (Yidana and Yidana, 2010; Mallick *et al.*, 2018; Singh *et al.*, 2017; Singh *et al.*, 2011; Machiwal and Jha, 2015) geochemical modeling (Ledesma-Ruiz *et al.*, 2015; Suma *et al.*, 2015; Singh *et al.*, 2013; Yidana *et al.*, 2008; Mallick *et al.*, 2018), stable isotopes (Barbieri *et al.*, 2005; Carucci *et al.*, 2012; Mallick *et al.*, 2018), redox indicator (Mallick *et al.*, 2018), structural equation modeling (Belkhiri and Narany, 2015; Mallick *et al.*, 2018) are currently employed as crucial techniques for understanding geochemistry and hydrogeochemical processes. Understanding this hydrogeochemical processes helps to determine a good quality of groundwater for the inhabitants. In the present study, hydrochemical analysis, spatial distribution of electrical conductivity, rock-water interaction, trilinear plotting to visualize the chemical composition of water, saturation indices, bonding between the chief ions has been studied. The findings of the present study make it simple to comprehend the evolution of hydrogeochemical properties.

Study Area

The study area is in the district of Kanniyakumari of the Tamil Nadu state of India. The Kuzhithuraiyar River and its principal tributaries, the Kodayar and Paraliyar Rivers, drain the area primarily. Since the research area is close to the equator, it is challenging to identify the summer season, and it has a tropical
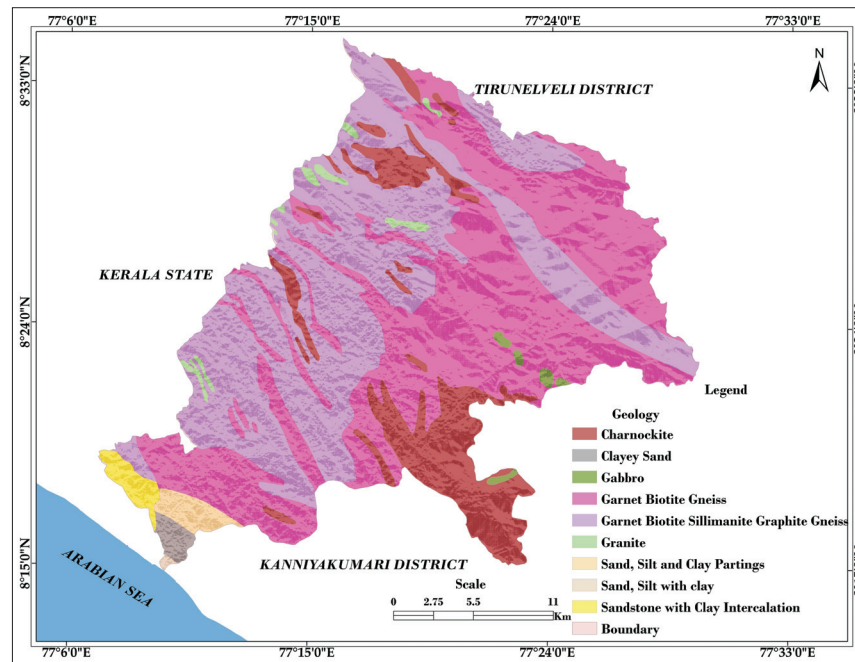
Fig.1. Geology map of Kuzhithuraiyar Sub-Basin

environment with an average temperature of 26.9°C. It experiences both the North East (NE) monsoon (October–December) and the South West (SW) monsoon (June–September) (Raxana and Venkateswaran, 2023). The most popular groundwater abstraction devices for irrigation in the research area are dug wells.

Geological Setting

The Kuzhithuraiyar Sub-Basin belongs to Southern Granulitic terrain which has a heterogeneous geology. Crystalline rocks of Archean to Proterozoic age occupy 97% of the area while rest of the area is covered by Miocene to Pliocene sediments and sedimentary rocks. The majority of the study area's northern and central regions are made up of garnet biotite gneiss, garnet biotite sillimanite gneiss, and garnet biotite graphite gneiss, with sporadic patches of charnockite, gabbro, and granite. Southeastern part is covered by Charnockite. The Southwestern part of the area is covered by clayey sand, Sand, Silt and Clay Partings, Sand Silt with Clay and Sandstone with Clayey Intercalations (Fig. 1; Kaliraj *et al.*, 2015; Raxana, 2023; Raxana *et al.*, 2024).

Hydrogeology

Formations that are both porous and fissured are found underneath the research region. The significant aquifer system in the studied region is composed of unconsolidated and semi-consolidated formations as well as worn, fissured, and fractured crystalline rocks. In the worn, fissured, and fractured crystalline rocks as well as in the sand dunes close to where the Kuzhithuraiyar River meets the Arabian Sea, groundwater occurs in phreatic conditions. The nature of the aquifer in the Kuzhithuraiyar Sub-Basin is confined to semi-confined. At a depth of 8 to 18 m bgl, the water table is found in the worn and semi-restricted to confined conditions in the fracture and fissured zones of these rocks. Water levels in sand dunes range from 4 to 8 m below ground level (Raxana *et al.*, 2024).

Materials and Methods

Sampling and Analysis

Using ArcGIS 10.5, the study region was partitioned into a number of 3km by 3km grids. The basin's 48 spots were chosen at regular intervals. Samples were collected in accessible areas of hilly regions, plain and coastal regions. Standard protocols for collection and analysis were followed, and 48 samples were collected during pre monsoon period. In the lab, the following variables were examined: pH, EC, TDS, and chief ions Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, SO_4^-, CO_3^-, HCO_3^-, and NO_3^-. An ions electrode was used to detect pH and EC. EC was multiplied by 0.64 to get TDS. Cations like Ca^{2+} and Mg^{2+} as well as anions like Cl^-, CO_3^- and HCO_3^- were analyzed using the titrimetric method. Na^+ and K^+ were examined using a flame photometer, and SO_4^- and NO_3^- were examined using a spectrophotometer. After all samples were analyzed, the normalized charge balance index (NCBI) was calculated using the following formula: (Edmond *et al.*, 1995; Huh *et. al.*, 1998; Mallick *et al.*, 2018).

$$NCBI = \frac{\Sigma T_z^- - \Sigma T_z^+}{\Sigma T_z^- + \Sigma T_z^+}$$

Where ΣT_z^+ is the total sum of cations in epm and ΣT_z^+ is the total sum of anions in epm

Results and Discussion

Hydro-geochemistry

Surface water seeping into aquifers is regarded as groundwater. The geochemical constituents of surface water also contribute to our understanding of how the quality of water changes over time (Mostafa, *et al.*, 2017). The outcome of Hydro-geochemistry reveals that the pH values (Fig.2) in the Kuzhithuraiyar Sub-Basin range from 8.5 to 6.5, with an average of 7.7. Ec value ranges from 37.50 µS/cm to 1996 µS/cm, with an
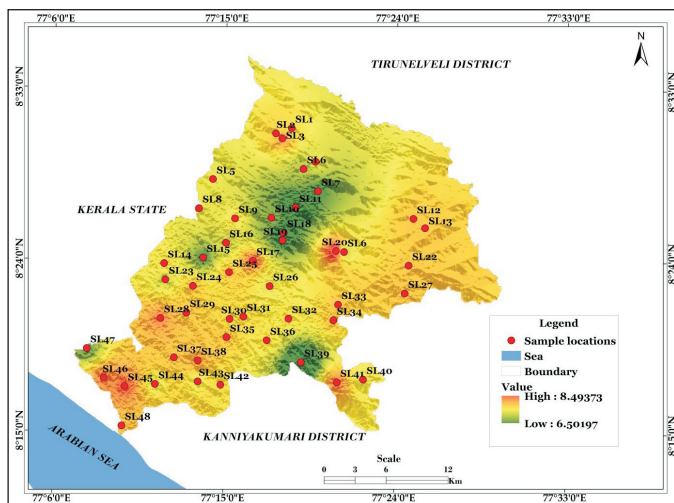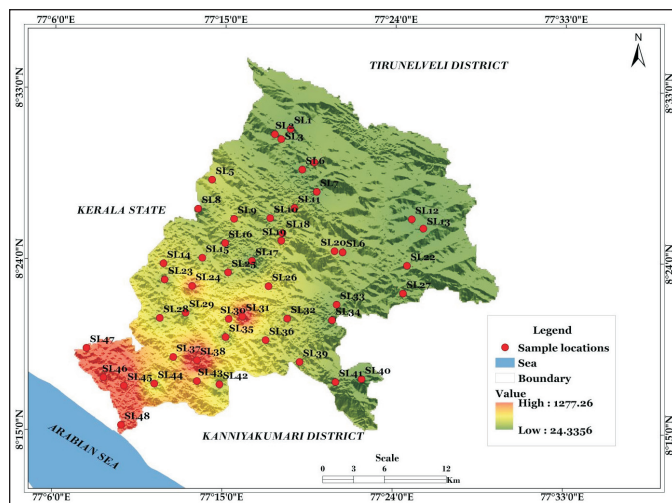
Fig.2. Spatial distribution map of pH

Fig.3. Spatial distribution map of TDS

average of 441.53 S/cm. TDS values (Fig.3) ranges from 24 to 1277mg/l. Following is a trend for cations and anions: Na >Ca> Mg > K and Cl> HCO_3> SO_4> NO_3> CO_3. With an average of 62.12 mg/l, sodium levels range from 317 mg/l to 3 mg/l. The average calcium level is 22.04 mg/l, with values between 92 mg/l and 2 mg/l. The average magnesium level is 12.22 mg/l, with values ranging from 58.32 mg/l to 1.22 mg/l. With an average of 6.82 mg/l, potassium levels range from 37 mg/l to 1 mg/l. The average chloride value is 94.26 mg/l, with values ranging from 624 mg/l to 4 mg/l. With an average of 90.96 mg/l, bicarbonate concentrations range from 366 mg/l to 4.97 mg/l. Sulphate has a concentration range of 77 to 1 mg/l, with an average of 15.76 mg/l. Nitrate concentrations range from 18 mg/l to 0.05 mg/l on average. With an average of 0.42mg/l, carbonate has a range of 0 to 4.33mg/l.

Distribution of Electrical Conductivity

Electrical resistivity is the reciprocal of electrical conductivity, which is the capacity to transfer electric current (Thompson *et al.*, 2018). The influence that TDS and EC have on a water sample's corrosivity as well as the solubility of marginally soluble chemicals like $CaCO_3$ make them significant (Nas and Berktay, 2010; Vincy, *et al.*, 2015). Water chemistry is indirectly impacted by the sample's electrical conductivity. Ec can be divided into four different saline concentration categories: low (250μS/cm), medium (250-750μS/cm), medium-high (750-1250μS/cm), and high (>1250μS/cm) (Fig.4). There are five high saline samples, 19 low saline samples, 22 medium saline samples, 2 medium to high saline samples. The Kuzhithuraiyar Sub-Basin has high electrical conductivity on its south western side, which is close to the coast which implies the influence of seawater intrusion and anthropogenic activities among the samples. The low electrical conductivity on its north eastern sides indicates that the area is having dense vegetation, non-contaminated clean and fresh water. The central part of the sub basin shows medium electrical conductivity due to interaction of rock and water for a longer time.

Rock-Water Interaction

Many chemical processes such as dissolution/precipitation, ion exchange processes, oxidation and reduction occur during rock-

water interaction (Dehnavi *et al.*, 2011). Gibbs plot gives an understanding about the water samples covering three different kind environments namely evaporation dominating area, rock dominating area and precipitation dominating area (Fig.5). The Gibbs plot reveals that most samples fall within the rock dominance zone, whereas only a small number of samples fall within the precipitation dominance zone and two samples fall within the evaporation crystallization zone. Rock dominance indicates that as water percolates, rock-water interaction affects the chemistry of the water. Most of the samples in central part of the study area have more residence time hence rock water interaction is more in these regions. In the hilly terrain the slope is steep, residence time of water is less and the water in these regions would be younger due to frequent precipitation. Hence, the samples in the hilly region show less TDS. Two areas with high TDS fall under evaporation zone, which means samples near the coastal region are, subjected to high evaporation as a result the chemistry of the groundwater changes by increasing salinity.

Trilinear Diagram

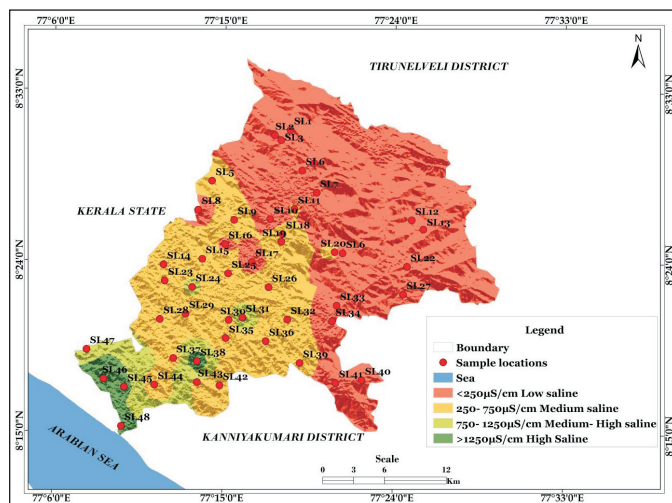Pipers trilinear plot introduced by Piper (1945), is realistic

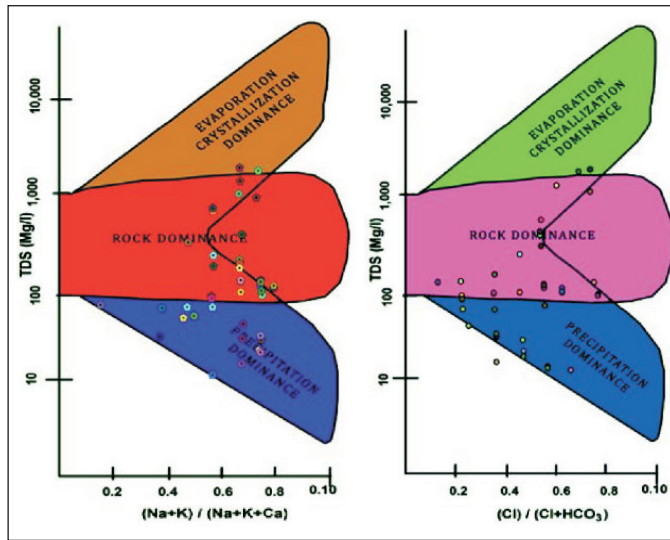**Fig.4.** Sample location and Spatial distribution map of EC (μS/cm)

Fig.5. Gibbs plot of Chemical Parametres of groundwater of Kuzhithuraiyar Sub-Basin
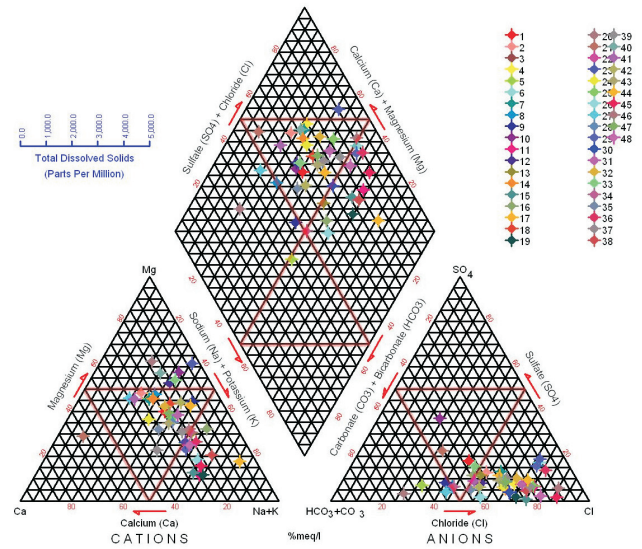

Fig.6. Pipers trilinear plot of Chemical Parametres of groundwater Kuzhithuraiyar Sub-Basin

for copious purposes which includes understanding the hydrochemical facies (Karanth, 1991; Sadashivaiah *et al.*, 2008; Adithya *et al.*, 2016), classifying the water types (Wen *et al.*, 2005; Adithya *et al.*, 2016), determining the geochemistry of hardrock (Srinivasamoorthy *et al.* 2008; Adithya *et al.*, 2016), identifying the hydrochemicalfacies (Chidambaram *et al.*, 2008; Adithya *et al.*, 2016), identifying the processes involved in groundwater recharge (Thivya *et al.*, 2013; Adithya *et al.*, 2016).

Ca-Na-Cl type, Na-Cl type, Mg-HCO$_3$ type samples constitute the majority of the samples (Fig. 6). Ca-Na-Cl and Na-Cl type of water indicates that strong acids exceed weak acids in the water samples. The Chloride ion in water may be due to the use of fertilizers for agriculture or due to evaporation process. In Mg-HCO$_3$type of water specifies weak acids exceed strong acids and dissolution of dolomite.

Saturation Index

If saturation index exceeds that of than 0, it implies that particular mineral is not completely dissolving in the groundwater. If the saturation index is less than 0, the groundwater is oversaturated with respect to that mineral and cannot dissolve the majority of it (Rina *et al.*, 2013). Using Phreeqc Interactive software, the saturation indices of a few minerals based on main ions were calculated. The carbonate group of minerals include calcite (CaCO$_3$), aragonite (CaCO$_3$), and dolomite (CaMg(CO$_2$)$_2$) containing Ca^{2+} and HCO$_3^-$ (Fig. 7a). Since there is oversaturation, it can be assumed that groundwater precipitated the minerals. The relationship between the saturation indices of calcium-bearing minerals and the SO$_4$ content in groundwater shows saturation of anhydrite (CaSO$_4$) and gypsum (CaSO$_4$.2H$_2$O) to the samples adjacent to the coast due to high evaporation process than other plain and hilly regions (Fig. 7b). Halite and a few other salt minerals dissolve in the presence of under-saturated water due to their high solubility. This is typical of non-marine habitats and typically occurs in the phreatic zone as a result of the movement of meteoric groundwater (Warren and Warren, 2016). The correlation between the halite saturation index and the level of chloride in the water samples demonstrates under-saturation (Fig. 7c), which suggests

that the breakdown of halite along the flow channel increases the concentration of sodium, chloride, and TDS.

Bond Between Chief Ions

Halite dissolution is represented by the Na *vs.* Cl. relationship (Fig. 8a). A higher concentration of Na than Cl indicates that sodium is liberated during the weathering of silicates. It suggests that most
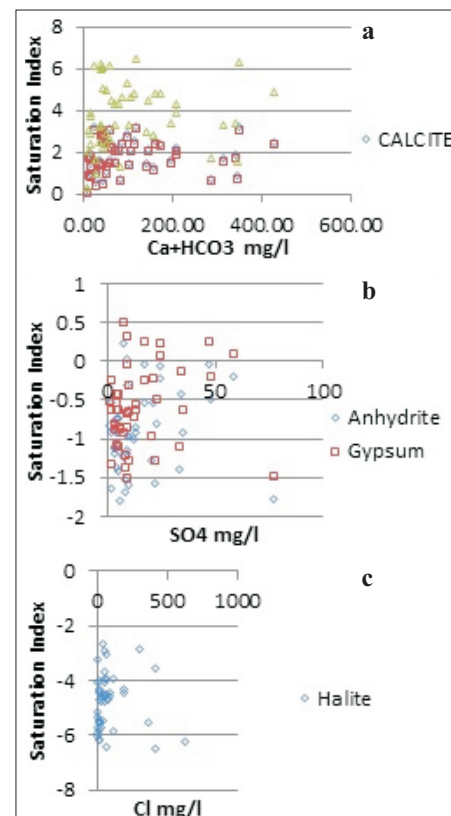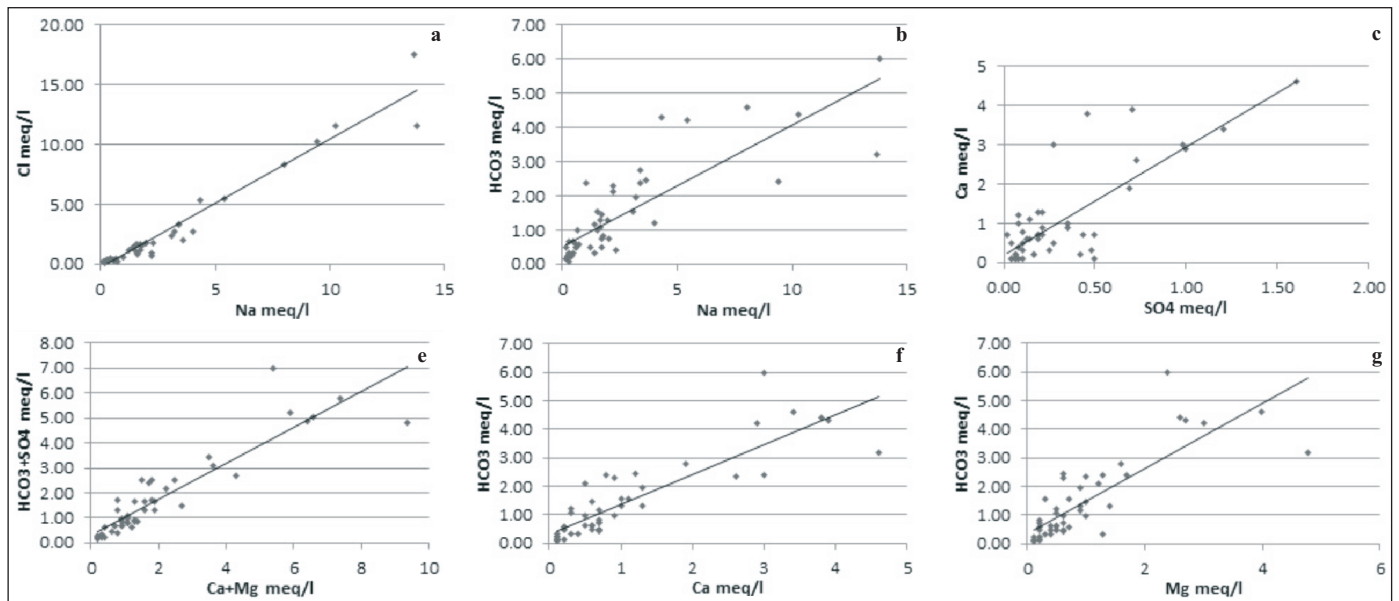

Fig.7. a) Plots of Ca+HCO$_3$ vs saturation indices of carbonate minerals, b) Plots of SO$_4$ vs saturation indices of CaSO$_4$ minerals, c) Plots of Cl- vs saturation indices of NaCl minerals

Fig.8. Scatter plot showing the correlation of major cations/anions to discriminate the geochemical processes **a)** Na^{2+} *vs* Cl^- , **b)** Na^{2+} *vs* HCO_3^- , **c)** SO_4^- *vs* Ca^{2+} , **d)** $Ca^{2+}+Mg^{2+}$ *vs* $HCO_3^-+SO_4^-$, **e)** Ca^{2+} *vs* HCO_3^- , **f)** Mg^{2+} *vs* HCO_3^-

samples are weathered by silicate and that only a small number of samples near the equiline are dissolved by halite. Silicate weathering results in an increase in HCO_3^- concentration. Na *vs*. HCO_3^- scatter plot (Fig. 8b) and Ca^{2+} *vs.* SO_4^- relationship (Fig. 8c) demonstrates that most samples are above the equiline and few samples are below the trend line. The link between $Ca^{2+}+Mg^{2+}$ *vs* $HCO_3^-+SO_4^-$ indicates both carbonate and silicate weathering (Fig. 8d; Datta and Tyagi, 1996). The samples positions above, below, and along the equiline indicate weathering caused solely by carbonate, weathering caused solely by silicate, and weathering caused by both carbonate and silicate. The Ca^{2+} to HCO_3^- ratio map (Fig. 8e) shows that the concentration of bicarbonate is higher than that of calcium, indicating weathering of anorthite and dolomite. Samples along the equilibrium line show gypsum dissolution, samples above the equilibrium line show excess calcium ions, and samples below the equilibrium line show excess bicarbonate. The relationship between Mg and HCO_3^- (Fig. 8f) indicates the source of dolomite along with dissolution of biotite.

Conclusions

The geologically heterogeneous nature of the study area indicates different processes in the interaction between rocks and water. Gibbs diagrams show that most of the samples fall under rock dominance, indicating that the rock-water interface is an important factor driving geochemistry. Samples near to the coastal tract are subjected to evaporation resulting in high salinity whereas samples in the plain and hilly region show low salinity due to rock dominance and precipitation. Major cation/anion correlation

diagrams distinguish geochemical processes. Most samples undergo silicate weathering, some undergo carbonate weathering, but few undergo both silicate and carbonate weathering. The average precipitation in the study area is 1600 mm, which causes the dissolution of rock salt present in the soil zone and increases the sodium concentration. The calcium-to-sulphate ratio indicates transfer of sulphate by fertilizers used in agriculture, with few samples showing decomposition of calcite, dolomite and anorthite along with gypsum. Thus, in general, the main processes driving water chemistry in the study area are silicate weathering, basic ion exchange, reverse ion exchange, recharge water, and dissolution processes.

Authors' Contributions

AGR: Conceived and Designed the Manuscript, Conducted the Experiment. **VS**: Reviewed and Edited the Manuscript.

Conflict of Interest

The authors declare no conflict of interest.

Acknowledgements

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