

Geochemistry of Groundwater in The Sedimentary Terrain around Devak and Rui Watersheds, Jammu and Kashmir, India



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Abstract

Groundwater quality survey has been carried out in the sedimentary terrain of Devak and Rui watershed, Jammu and Kashmir, India. The area enjoys sub-tropical to moist temperate climate with the average temperature varying between 20 C -20⁰ C in winter and 33⁰ C -47⁰ C in summer. The sedimentary rock includes the sandstone, mudstone, shale, silt and clay. The WATEQ2 computer programmer was used to compute the ionic activities, using the Debye - Huckel and Davis equation and the mineral saturation index (SI) for CaCO₃. The water chemistry dominance of Ca-HCO₃, indicating recharging water in sandstone aquifer suggesting the rock dominance. The average silica activity is 10^{-3.87}. Results of the work indicates that the geochemistry of groundwater in the terrain is probably, controlled by the incongruent processes and dissolution of carbonate minerals that control the chemistry of groundwater. Kaolinite is the stable phase. The ability of soil to take up the ions during the pre-monsoon period results from the formation of new clay minerals and precipitate of calcite due to the leaching from the soils occurred to a greater extent in the post-monsoon period.

Keywords: Groundwater; geochemistry; sedimentary terrain; Devak and Rui, Watersheds; Jammu and Kashmir.

Introduction

Groundwater geochemistry has potential use for tracing the origin and water composition change. Introduction Groundwater geochemistry has potential use for tracing the origin and water composition change through reactions with the environment in which the water is circulated. There is great need in present day for the hydrologist to obtain the informations concerning the residence times, flow paths and aquifer characteristics which are often influenced by human activities. The chemical composition of groundwater is also determined by a number of processes, which include atmospheric input, interaction of water with soil and rock and input of chemicals derived from human activities. It is not easy to separate the impact of natural weathering and by anthropogenic input on the chemistry of groundwater. The accurate simulation of aqueous geochemistry through the use of computer models can serve as an invaluable aid in characterizing water quality and its availability as well in determining mineralogical controls on natural water chemistry. If the controlling influences of an aquifer system is determined, the effect of man made can be evaluated and plan to minimize deleterious effect can be considered.

In the last a few decades, there has been an incredible increase in the demand for freshwater resource due to rapid population growth and industrialization (Li et al. 2012; Jasrotia and Kumar 2014a; Jasrotia et al., 2016; Li and Qian 2018; Adimalla and Li 2018; Jasrotia et al. 2018; Jasrotia et al. 2019). Therefore, a number of researchers focused on groundwater quality in different regions. Krishna Kumar et al. (2014) conducted a study on groundwater quality in and around Vedaraniyam, South India, and found that the groundwater was contaminated by acute usages of pesticides in agricultural regions. El Alfy et al. (2015) extensively studied on groundwater quality and its pollution assessment using multivariate geostatistical techniques in arid areas, Saudi Arabia, and noticed that dissolution of various minerals, evaporation, and human impact on the

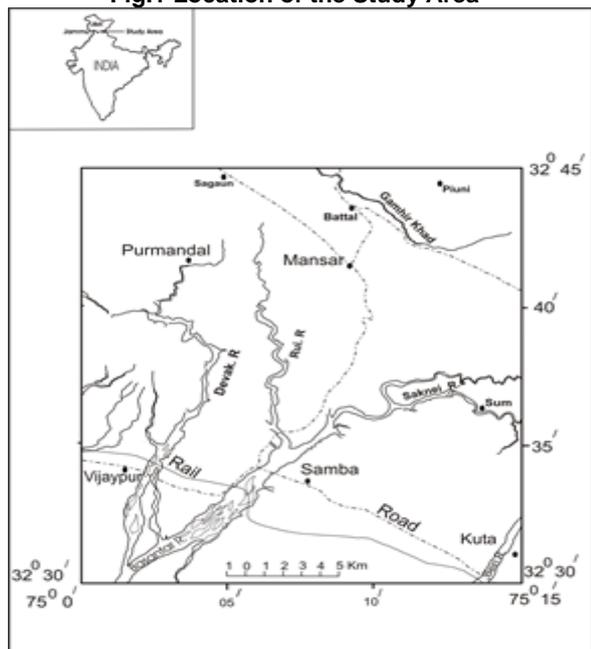
aquifers deteriorated the groundwater quality. Ayadi et al. (2018) investigated the geochemical assessment of groundwater quality in Northwestern Tunisia.

Moreover, hydrogeochemical characteristics and assessment of groundwater quality using GIS technique have been carried out by many workers (Jasrotia et al. 2012; Krishna et al. 2015), because it is one of the prominent methods to delineate the groundwater quality for various purposes. Groundwater quality distribution maps may be used to assist planners, managers, and local officials in evaluating the potential of contamination and as precautionary indication of hazardous zones. GIS has emerged as a dominant tool for accumulating, analyzing, and displaying spatial data, and these data were used for decision making in several areas including geological and geo-environmental fields (Jasrotia and Kumar 2014b). As the study area is being developed and the agricultural land converted into non agricultural uses with its deleterious effects on groundwater quality, it is essential to know the geochemistry of the groundwater of the study area. For the proper planning and development of the study area a detailed groundwater quality analysis with respect to the hydrochemistry, requirement of waters used for domestic, Irrigation and industrial purposes is essential. The present study describes the chemical characteristics of groundwater mostly controlled by the incongruent processes and dissolution of carbonate minerals that control the chemistry of groundwater

Study Area

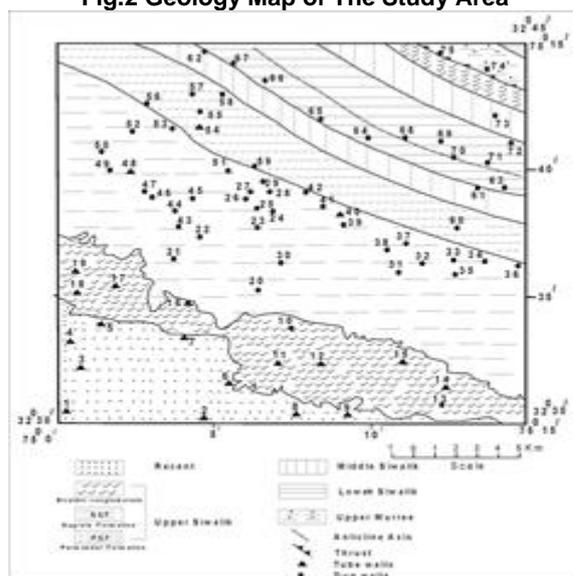
The present study area around Devak-Rui watershed lies between latitude 32° 30' N to 32° 45' N and longitude 75° 0' to 75° 15' E and geographical coverage of the area of 650 Km² (Fig. 1) with dense populated, come under the urban as well as rural developing areas.

Fig.1 Location of the Study Area



The average rainfall estimated from the number of years of the study area 1116 mm of which monsoon rainfall is 800 mm. The area falls under the sub-tropical to moist temperate climate conditions in which the average temperature of 2°C - 20°C in winter to 33°C- 47°C in summer. Physiographical the study area has low to moderately high linear ridges with moderate to steep slope with varying in height from 400m to 1400m. The structure and lithology have played a major role in the evolution of geomorphology in the hilly area. The strike of bedding has controlled the trend of the hill range to a large extent. The strike and dip of the rock formation, joints and major tectonic elements have played significant roles in the development of drainage pattern. Geomorphologically the study area investigated can be broadly divided into four geomorphic units such as Structural hills, Piedmont alluvial plain, Older alluvial surface and Present day flood Plain. The geological formation mainly comprises Upper Murree and Siwalik rocks, exposed in uplifted thrust sheets which record Early Miocene through Pleistocene synorogenic foreland basin sedimentation. However, on the basis of lithology the study area has been divided into Upper Murree, Lower, Middle, Upper Siwalik Subgroup and alluvium of Jammu formation (Fig 2.).

Fig.2 Geology Map of The Study Area



Methodology

Groundwater samples from Seventy-Five Dug wells and Tube wells from the different part of the study area were collected during Pre-Monsoon season and post –monsoon for seasonal variations and were analyzed their chemistry. The samples collected are extensively used for drinking and other domestic purposes. The techniques and methods for collection and analysis for water samples followed standard water quality procedure, American Publication Health Associations (1992) and Manual of Pollution Control Board (1997). pH, EC and TDS measured at the site by using pH, conductivity and TDS testers. The sodium (Na⁺) and potassium (K⁺) were determined by Flame Photometer. Total hardness (TH) as CaCo₃, total alkalinity (TA) as

CaCO₃. The D.O analyzed in the laboratory by Winkler titrimetric azide modification (iodometric) method. Calcium (Ca²⁺), bicarbonate (HCO₃⁻) and Cl⁻ were analyzed by volumetric method. Magnesium was calculated from TH and Calcium (Ca²⁺) contents. The Fluoride (F⁻) was analyzed by Fluoride Ion selective electrode. The sulphate (SO₄²⁻) Nitrate (NO₃⁻) and Silica (SiO₂) were determined by the UV spectrometer photometric. All the concentrations are expressed in milligram per little (mg/l) expect pH and EC in μmhos/l.

Water Chemistry

The computer program WATEQ2 was used to compute the equilibrium distribution of aqueous species and then calculate mineral saturation indices (Ball, 1980). This PL/I code is a revised and expanded version of WATEQ the computerized aqueous chemical model of Truesdell and Jones (1974). WATEQ2 computer programmer was used to compute ionic activities such as options are provided for calculation of activity coefficient using either the Debye - Huckel or Davis equation and the mineral saturation index (SI) for CaCO₃

$$SI = K_{IAP} / K_{SP}$$

Where K_{IAP} is the ionic activity product and K_{SP} is solubility product of CaCO₃ (10^{-8.35}) value from Kraus Kopf (1979). A positive SI indicate precipitation, a negative SI dissolution and a natural SI an equilibrium state for the solid phase of CaCO₃. The activities of aqueous solution have been used to calculate the saturation state of mineral. If a solution is under saturated with respect to a mineral the mineral tend to dissolved; If a solution is oversaturated with respect to a mineral the mineral tend to precipitate. Saturation with respect to the mineral indicated that solution and mineral are in equilibrium. The solution concentration CaCO₃ shows that all samples having positive value more than zero (Table 2) indicate the mineral tend to precipitation due to oversaturated solution.

Piper^s Trilinear Diagram

In the Piper^s Trilinear diagram (Fig.3) plots of the hydro chemical data of the groundwater samples of the study area shows the dominance of calcium bicarbonate, that all the samples falls in the field 1,3 and 4 which suggest that alkaline earth exceeds alkalies and weak acids exceeds strong acids respectively. The Ca, Mg and HCO₃ indicate the carbonate hardness that is total hydrochemistry is dominated by alkaline earth and weak acids.

Expanded Durov Diagram

In the Expanded Durov Diagram (Fig.4) groundwater samples of the pre monsoon and post monsoon from the production wells shows that majority of samples falls in the field 1, which provides a distinct classification of the combination of dominant cations and anions such as Ca- HCO₃ dominance, indicating recharging water in sandstone aquifer and rest of the samples fall in the other fields 2 and 3. The field 2 indicating that groundwater is Ca- SO₄ water is associated with sedimentary rocks. The deposition of Ca- SO₄ from the saturated solution is also induced by a rise in the Ca- HCO₃ content in consequence of raised CO₂ pressure, which causes a rise in the content of Ca²⁺.

Stiff Diagram

Stiff diagram for representing hydro chemical data of the groundwater samples by four parallel axes as shown in (Fig. 5). Concentration of cations are plotted to the left of a vertical axis and anions to the right in the stiff diagram shows gradual uniform increase of calcium and bicarbonate whereas sodium plus potassium, chloride, magnesium and sulphate decrease in concentration.

Gibb Diagram

Gibbs (1970) proposed three controlling factor dominance water chemistry such as evaporation, rocks and precipitation. All the samples fall in the central portion of the Gibb diagram (Fig. 6) suggesting rock dominance over the chemistry of groundwater. It caused by the interaction between chemistry of aquifer rock and percolating precipitated water in the sub-surface.

Fig.3 Piper Trilinear diagram

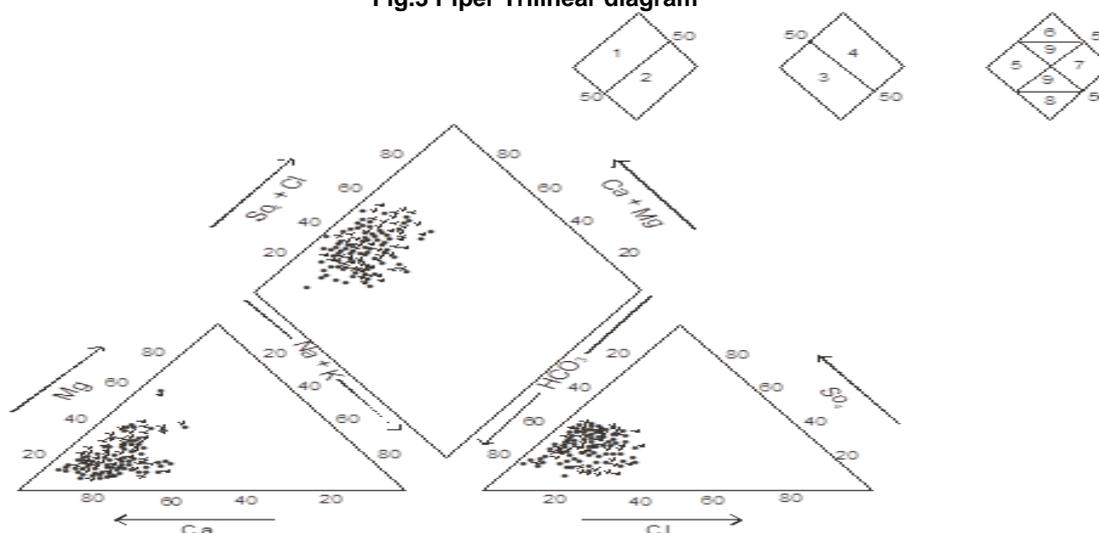


Fig.4 Expanded Durov diagram

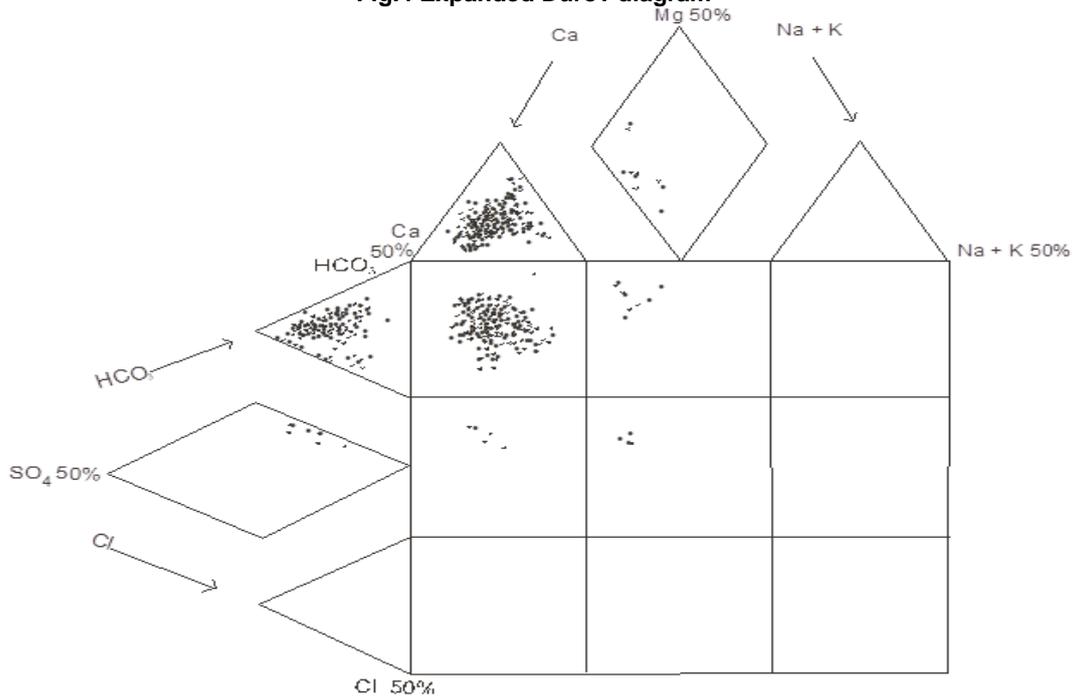


Fig.5 Stiff diagram of the groundwater quality

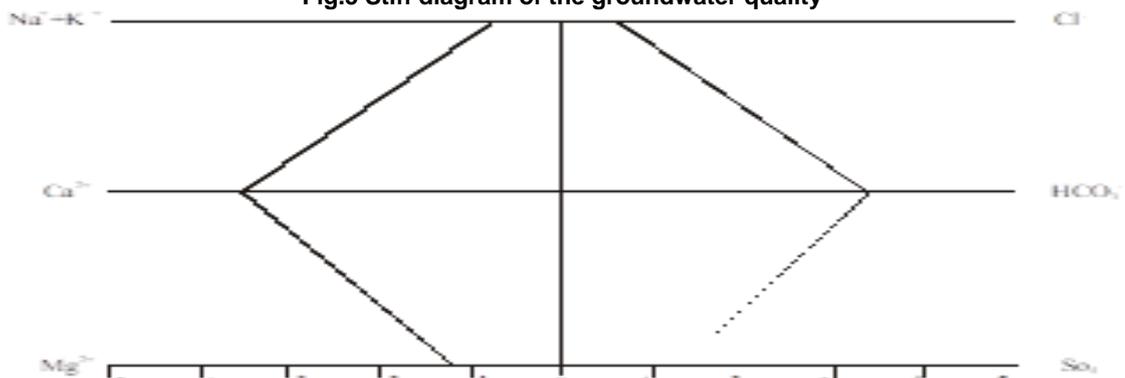
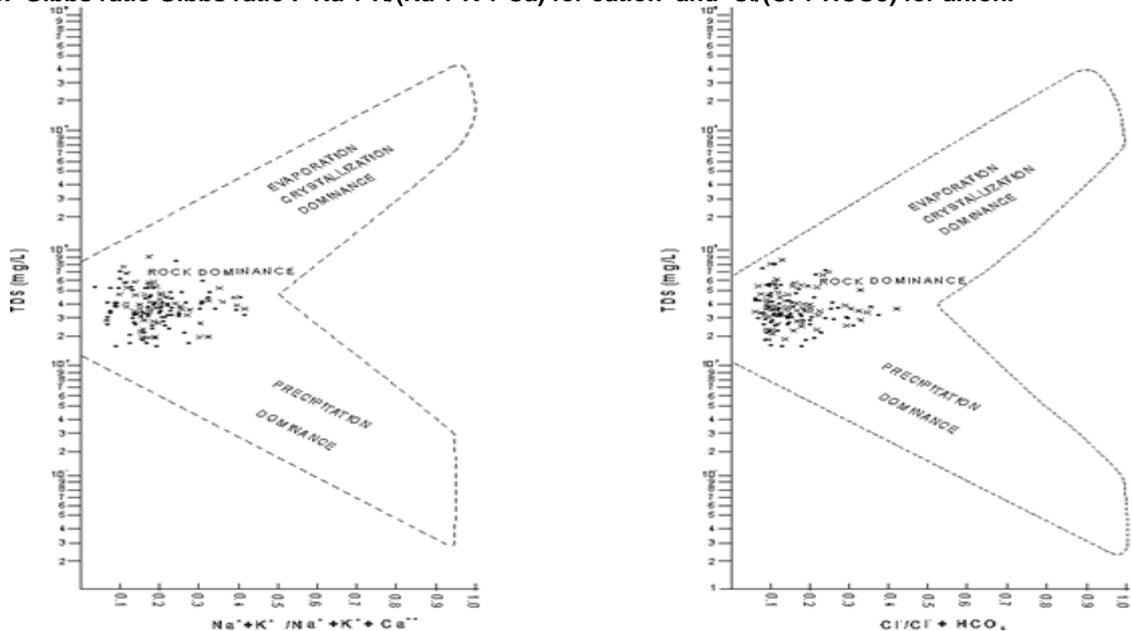


Fig. 6. Gibbs ratio $1 - \frac{Na + K}{Na + K + Ca}$ for cation and $\frac{Cl}{Cl + HCO_3}$ for anion.



Water Quality

The groundwater quality of the study area is compared with water quality standard prescribed by Indian standard specification (Table 1.) The groundwater has TDS, TH, Ca, Mg, HCO₃ and NO₃ shows concentration in excess of save limit, i.e. 500, 300, 74, 30, 200 and 45 mg/l, which can effect human health in such a way. Water with high TDS having more ionic concentration, which is reduce the potability and can induce an unfavorable physiological reaction in the consumers. The TH varies from 90.6 mg/l to 542 mg/l indicate the groundwater of the study area moderate hard to very hard, which is permanent in nature caused by the dominance of week acids (TA<TH). The hardness cannot be removed easily by the boiling water. Excess concentration of TH has no adverse effect on health, but it prevents the formation of lather with soap and increase the boiling point of water. The Cl concentration generally imparts a salty taste to water and salty taste indicates the pollution. Those who are not accustomed to high concentration of Cl in water, it may cause a laxative effect. High concentration in NO₃ in groundwater causes the blue baby in children. WHO (1984) has reported that 80% of the health hazards occur in the world, because of consumption of inferior water quality. The groundwater is suitable for irrigation because the EC value in the groundwater in the study area ranges from 260µS/cm to 1290µS /cm at 25°C of pre-monsoon period and 290µS/cm to 1360µS/cm at 25°C of post-monsoon period. Classified groundwater for irrigation suggested by Wilcox (1967) according to EC values, the groundwater of the study area is of good to permissible limit. According (Doneen 1964; Domenico and Schwartz, 1990) which implies that the groundwater is of good quality for irrigation purposes as per as the permeability index is concerned. According to Industrial Water Quality Standards (American water quality works Association, 1971) the groundwater in study area is suitable for Industrial purposes where all samples shows total hardness (TH) and TDS within the permissible limit except two cases. The industrial sector in India is incurring a loss of about 200 crores of rupees per year through adverse chemical reaction (Jain, 2000).

Mineral Water Interactions

Water percolating has a corrosive action due to the dissolved CO₂ present and the primary source of CO₂ dissolved in natural water is the earth atmosphere. Groundwater has additional supply from decay of organic matter in the soil zone. The result is a mildly acidic solution of carbonic acid which acts as a solvent for minerals. The carbon dioxide from the carbonic acid which is neutralized in reactions with the minerals in ground water. The silicate yield cations and silica are transformed to secondary clay mineral. The news minerals formed in sedimentary rocks include Kaolinite, montmorillinite, chlorite and illite. The reaction continues till all the minerals in the stage of equilibrium with water.

To illustrate weathering reactions, it is conventions to use stability diagram constructed by means of the thermodynamics data for minerals. Stability diagram have been widely used for understanding the geochemical behavior between mineral and water phase. With thermodynamic data from Helgeson (1969) and average silica activity of 10^{-3.87} the stabilities of illite, Kaolinite, Montmorillinite and Chlorite shows that Kaolinite is the stable phase (Fig. 7).

Fig.7 : Mineral stability diagram of the groundwater samples

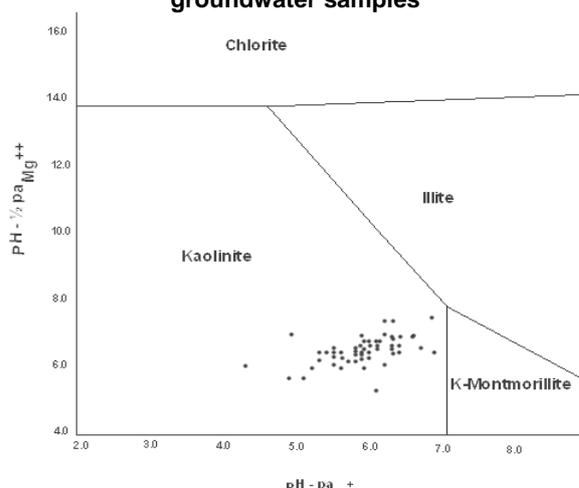


Table 1. Comparison of the Chemical Composition Groundwater of the Study Area with Indian Standard Specification for Potable water

S. No	Water Quality Parameters (Units)	Indian Standard Specification		Chemical Concentration
		Desirable Limit	Permissible Limit	
1	pH	6.5-8.5	No relaxation	6.5 - 8.3
2.	TDS (Mg/l)	500	2000	170-830
3.	Calcium (Mg/l)	75	200	20-152
4.	Magnesium (Mg/l)	30	100	2- 80
5.	Potassium (Mg/l)	No limit specified		0.05 -28
6.	Sodium (Mg/l)	No limit specified		3 - 45
7.	Bicarbonate (Mg/l)	200	600	80-450
8.	Chloride (Mg/l)	250	1000	2-120
9.	Sulphate (Mg/l)	200	400	15-110
10.	Nitrate (Mg/l)	45	100	2-55
11.	Fluoride (Mg)	1.0	1.5	.02-0.47
12.	Total Hardness as CaCo ₃	300	600	90.6-542

Fig.8 Thermodynamic stability plot for (a) Ca, (b) Mg system, (c) Na and (d) K system

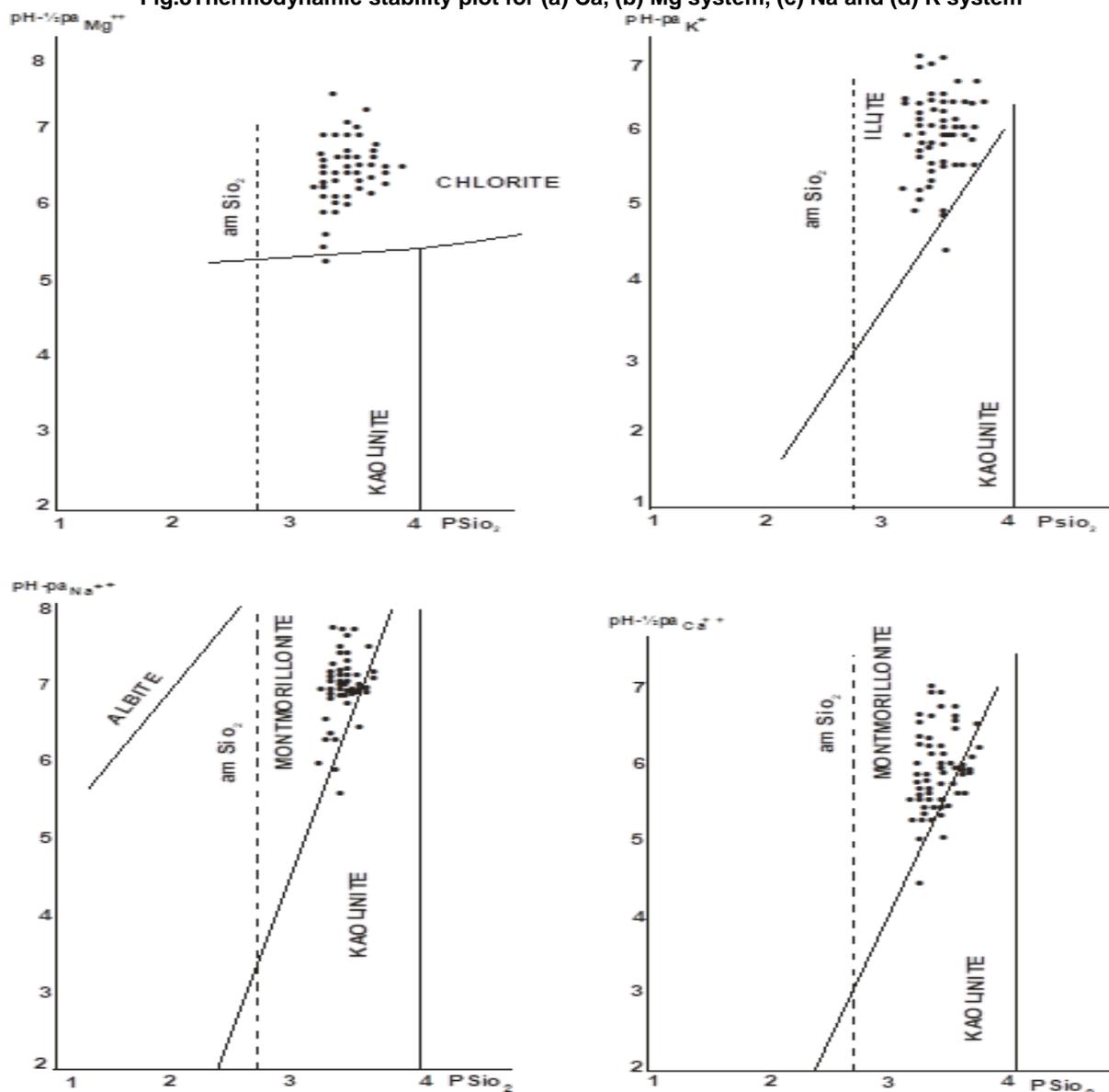


Table. 2. Geochemical parameters distribution

Parameters	Range
Na ⁺ : Ca ²⁺	0.06 to 0.95
Mg ²⁺ : Ca ²⁺	0.07- 0.86
Cl ⁻ : Na ⁺	0.12-4.86
Log (Ca+ Mg) / (Na) ²	(-0.32 - 0.97)
SI (CaCO ₃)	0.47-0.79
Index of Base Exchange (IBE)	
CA1	-7.71-0.757
CA11	-0.508-0.309

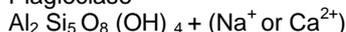
The stability diagrams have been prepared for understand the silicates of Ca²⁺, Mg²⁺, Na⁺, and K⁺, (Fig. 8) which were constructed earlier by Jacks (1973). The stabilities of minerals in these diagrams are represented as a function of composition of the water phase. Seventy five samples of the groundwater were analyzed for silica determination and these analyzed samples than plot in the stability

diagram using ion activities. The Debye-Huckel formula and mean salt method (Garrels and Christ, 1965) were used to determine the activities coefficients. In the stability diagram, samples are fall in, Na- montmorillonite, Ca- montmorillonite, chlorite and illite; it is evident that the mineral Na-montmorillonite, Ca- montmorillonite, illite, and chlorite should be stable. The water samples are generally found phase border between kaolinite and montmorillonite that is and due to the precipitation/leaching the concentration tends to increase the content among the cations and silica. The direction of transposition of the water composition on the plot is up towards to the left. Garrels (1976) stated that low concentration of Ca²⁺, Mg²⁺ and Na⁺ ions are associated with minerals, namely Kaolinite and quartz, whereas the abundance of these may be associated with the minerals like montmorillonite, illite,

and chlorite. Hence the formation of new clay minerals Na-montmorillinite, Ca- montmorillinite, illite and chlorite in the area is expected due to the excess supply of cations and silica to the pre-existing Kaolinite. The Na^+ and Ca^{2+} in the groundwater can be derived from the incongruent dissolution of plagioclase in sedimentary rocks.



Plagioclase



Kaolinite

Ca^{2+} comes from carbonate minerals such as calcite which commonly occur as cementing material in sedimentary rocks. The ability of soil and host rock to take up K, Na, Ca and Mg indicate that water regimen has been more leaching in the study area.

While the transforming of Kaolinite to other clay minerals described by the evaporation/evapotranspiration process, H^+ is released. H^+ combines with the HCO_3^- and form CO_2 . This may be one of the reasons for the high partial pressure CO_2 observed in the water than the atmospheric pressure. These waters pass through the soil decomposing the organic matter, which absorbs large amounts of CO_2 that is converted to HCO_3^- in the weathering reaction (Jacks, 1973). When the cations are removed HCO_3^- is thus corresponding anion can also be removed from the waters. The Ca^{2+} and HCO_3^- are thus removed from the water by means of precipitation of CaCO_3 . Consequently, all the groundwater samples in the study area found to have a positive Saturation index (0.47- 0.79) which suggest a precipitation of CaCO_3 .

According to the molar concentration Na^+ : Ca^{2+} ratios (Table.2) observed to be less than unity (0.06 to 0.95) except three cases indicating a Ca^{2+} concentration is more which might be result from the precipitation of CaCO_3 and ion exchange process. If the calcite precipitated from the solution containing an abundance of Mg^{2+} and Mg^{2+} could be further co-precipitated. Therefore, Ca^{2+} attains the higher concentration than Mg^{2+} in the groundwater and as a result the ratio of Mg^{2+} : Ca^{2+} is found to be less than unity (0.07-0.86) except one case in the study area. The log ratio of the molalities of exchangeable divalent and monovalent cations $\text{Log} (\text{Ca} + \text{Mg}) / (\text{Na})^2$ shows the progressive nature of ion exchange reactions. The negative to positive (-0.32 - 0.97) values of $\text{Log} (\text{Ca} + \text{Mg}) / (\text{Na})^2$ to identify the area and direction of flow in the aquifer system. The ion exchange process helps us to know the recharge and discharge of the areas by using the Scholler (1956) index of Base Exchange (IBE). The IBE shows negative to positive value in CA1 (-7.71- 0.75) and CA11 (-0.508-0.309) suggesting that exchange between Na and K in groundwater with Mg or Ca in rock/alluvium, both the indices are positive and if reverse both the indices are negative. More than 90 % of the areas shows the negative index indicating that host rock are the primary sources dissolved solids in the water.

Conclusion

The geochemistry of this system is probably controlled by the incongruent processes dissolution of carbonate minerals. The hydrochemistry of the groundwater in the study area dominance by Ca, Mg and HCO_3^- indicate the carbonate hardness that is total hydrochemistry is dominated by alkaline earth and weak acids. The ability of soil to take up the ions during the pre-monsoon periods resulting the formation of new clay minerals and precipitate of calcite. The concentration of ions increases in the post -monsoon because of recharge by leaching process and infiltration through the soil zone. The positive saturation index shows all the groundwater samples of the study area suggesting the precipitation of CaCO_3 . With thermodynamic data and average silica activity of $10^{-3.87}$ the stabilities of illite, Kaolinite, Montmorillinite and Chlorite shows that Kaolinite is the stable phase. The stability diagrams have been prepared for understand the silicates of Ca^{2+} , Mg^{2+} , Na^+ , and K^+ , majority of samples are fall in Na-montmorillinite, Ca- montmorillinite, chlorite and illite, it is evident that the mineral Na- montmorillinite, Ca-montmorillinite, illite and chlorite are stable.

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