JETIR.ORG ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

FLUORIDE-BEARING GROUNDWATER IN AND AROUND KESAMUDRAM MANDAL, MAHABUBABAD DISTRICT, TELANGANA STATE, INDIA

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Abstract

Groundwater samples were collected in parts of Kesamudram mandal, Mahabubabad district, Telangana State, India for two seasons (pre-monsoon and post-monsoon) and analyzed for chemical parameters with respect to fluoride. The fluoride concentration ranges from 0.52 to 3.40 mg/l and 0.08 to 2.60 mg/l during pre-monsoon (PRM) and post-monsoon (POM) seasons respectively. Results showed that water samples were contaminated with presence of fluoride ion. High fluoride concentration recorded during PRM 33% and POM 29% when compared with World Health Organization tolerance limit of 1.5 mg/l. Hydrogeochemical facies distribution indicates water-rock interaction resulted in occurrence of high fluoride concentration in groundwater. An analytical result plotted on Gibb’s diagram indicates rock and evaporation dominance. Principal component analysis indicates hydro-geochemical processes like weathering, ion exchange, and anthropogenic activities contribute to groundwater chemistry.

Keywords: Fluoride, Groundwater, Hydrogeochemical facies, Gibb’s plot and Principal Component Analysis (PCA)

Introduction

Fluoride contamination in drinking water due to natural and anthropogenic activities has been recognized as one of the major problems worldwide imposing a serious threat to human health. Fluorine has the highest electro-negativity and most reactive among all known elements. Fluoride ion (F^-) occurs in natural waters commonly in concentrations less than 1.0 mg/l and seldom outside the range from 0.01 to 10.0 mg/l. The amount of Fluoride ion (F^-) occurring naturally in groundwater is governed principally by climate, composition of the host rock, and hydrogeology. Some anthropogenic activities such as use of phosphatic fertilizers, pesticides and sewage and sludge, depletion of groundwater table, etc., for agriculture have also been indicated to cause an increase in Fluoride ion (F^-) concentration in groundwater (Ramanaiah et al. 2006). Due to excessive Fluoride ion (F^-) intake dental and skeletal fluorosis occur besides the muscle fiber degeneration low hemoglobin levels, excessive thirst, headache, skin rashes, nervousness and depression, etc. (Meenakshi 2006). In natural conditions the concentration of Fluoride ion (F^-) in water is generally <1 mg/l (Hem 1991). The (WHO, 2004) suggests that the highest desirable limit of Fluoride ion (F^-) is 1.5 mg/l in water for drinking purpose. In India, the Bureau of Indian Standards (BIS 2003) prescribed the Fluoride ion (F^-) limits 0.6 to 1.2 mg/l.

The reported literature on health hazards associated with long-term ingestion of F^- bearing water is summarized (Subba Rao 2012; Machendar et al. 2014). Around 200 million people from 25 nations suffer from ill-health because of a higher Fluoride ion (F^-) concentration in drinking water (Ayoob and Gupta 2006). In cold countries such as USA or UK, Fluoride ion (F^-) stricken fetal health problems are reported (Ayoob and Guta 2006). In Mexco, 5 million people (about 6% of the population) are affected by F^- in groundwater, while in Kenya and South Africa; the levels of Fluoride ion (F^-) exceed 25 mg/l (Grimaldo et al. 1997). In India, its concentration up to 38.5 mg/l is found (Susheela et al. 1999). The dreaded fluorosis prevalent in some parts of central and western China is a result of drinking of Fluoride ion (F^-) bearing groundwater. In India, about 60 million people (including 6 million children), especially in the states of Andhra Pradesh, Assam, Bihar, Delhi, Gujarat, Karnataka, Kerala, Madhya Pradesh, Maharashtra, Orissa, Punjab, Rajasthan, Talangana, Tamilnadu, and Uttar Pradesh, have no option except to drink Fluoride ion (F^-) contaminated drinking water (Mor et al. 2009; Raju et al. 2009 and Subba Rao 2011).

Many researchers worldwide including India studied the controlling factors of fluoride concentration in groundwater through geological, chemical and physical characteristics of aquifers, porosity of soil/rocks, climatic conditions, depth of water level and interaction of other chemical element for enhancement of fluoride distribution (Subba Rao 2011; Madhnure et al. 2007). But, these factors are not uniform in space and time. Thus, an examination of source of Fluoride ion (F^-) in groundwater over the country is a prerequisite before one can plan the strategies to mitigate the health problems. The chosen area for this study was, undertaken to investigate the quality of underground drinking water of Kesamudram mandal, Mahabubabad district is marked as one of the prevalent fluoride zones in Telangana State, India, this has prompted the authors to take up the present study to delineated fluoride vulnerable zones and to identify the geochemical process controlling the incidence of fluoride in groundwater.

Study Area

The study area covering about 148 sq. km falls in Kesamudram mandal, Mahabubabad district of Telangana State. Study area lies in between North Latitudes 17° 30' to 17° 45' and East Longitudes 79° 45' to 80° 00' (**Fig. 1**) and falls in the Survey of India toposheet No. 56 O/13 and 56 O/14. The study area receives average annual rainfall of 836.54 mm during the year 2010 to 2020 by southwest, northeast, winter and summer monsoons (**Table 1 and Fig. 1b**). The climate of the study area is generally hot. Average temperature in summer is 40^0C, in winter is 16^0C. The most of the study area was irrigated for paddy crops, turmeric, corn and red chilli were excessive use of fertilizers were common.

Geology

The major exposure of the study area is peninsular gneiss, and calc gneiss which have been marked for widespread fluoride-bearing minerals indicating their accessibility to water by weathering along with leaching process. Mica content is significantly noted in peninsular gneiss and apatite [$Ca_5 (PO_4)_3$ (F, Cl, OH)], hornblende Ca_2 (Mg, Fe, Al)$_5$ (Al, Si)$_8$ O_{22} $(OH)_2$ or biotite [K(Mg, Fe)$_3$ ($AlSi_3O_{10}$) (F, OH)$_2$] is noted which are the sources of fluoride into the groundwater (Purushotham et al. 2011).

Hydrogeology

The rocks in and around Kesamudram areas is referred to as granitic rock. Secondary porosity develops the rocks into aquifers. The weathered zone (characterized by low hydraulic conductivity) in the country rocks extends up to a depth of 25 m, whereas the fractured zone occurs up to a depth of 75 m from the ground surface. Groundwater exists under unconfined conditions in the weathered zone as well as under semi-unconfined conditions to confined conditions in the fractured zone. Groundwater is being exploited through open dug wells that tap the weathered zone, and bore wells are used to tap the fractured aquifer.

Materials and Methods

In order to assess the groundwater quality, twenty one groundwater samples have been collected in pre-cleaned polyethylene containers for pre and post-monsoon seasons. They were analyzed for pH, Electrical Conductivity (EC), Total Dissolved Solids (TDS), Total Hardness (TH), Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Sodium (Na^+), Potassium (K^+), Carbonate (CO_3^{2-}), Bicarbonate (HCO_3^-), Chloride (Cl^-), Sulphate (SO_4^{2-}), Nitrate (NO_3^-) and Fluoride (F^-) for all physico-chemical parameter using standard methods (APHA, 1995).

The pH was measured with Digital pH Meter (Model 802 Systronics) and Electrical Conductivity was measured with Conductivity Meter (Model 304 Systronics). Total Dissolved Solids were estimated by calculation method. Sodium and Potassium was measured with Flame photometer (Model Systronics 130). The TH (as $CaCO_3$) and Ca were analyzed volumetrically using standard EDTA. The concentration Mg is difference between the TH and Ca^{2+}. CO_3^{2-} and HCO_3^- were estimated by titrating with standard HCl using phenolphthalein and Methyl orange as acid-base indicators. The Cl^- was analyzed by titrating with standard $AgNO_3$. Sulphates and Nitrates were measured with Spectronics 21 (Model BAUSCH & LOMB). Fluoride concentration was measured with Orion ion analyzer with fluoride ion selective electrode. The units EC is expressed in micro-siemens per centimeter (µS/cm) at 25ºC, while those for the remaining chemical variables (except pH) are in milligrams per liter (mg/l).

Results and Discussion

The statistical parameters like minimum, maximum and mean concentration of physic-chemical parameters, major ion concentrations are tabulated in (**Table 2**).

Physico-chemical characteristics

Groundwater is from slightly acidic to alkaline nature, with pH varying between 7.36-8.56 and 6.76-8.0 during pre- and post-monsoon seasons. Total dissolved salts in the groundwater is between 205-2355 mg/l and 153-1730 mg/l in pre and post-monsoon seasons, high TDS is due to the influence of anthropogenic source, such as domestic sewage, septic tanks and agricultural activities (Krishna Kumar et al. 2017). The concentration HCO_3^- varies from 142-640 mg/l and 130-670 mg/l in pre and post-monsoon seasons. Bicarbonate, usually the primary anion in groundwater, is derived from the carbon dioxide released by the organic decomposition in the soil. The Cl^- range varies from 12-680 mg/l and 5-678 mg/l in pre and post-monsoon seasons. The Chloride in groundwater may be from diverse sources such as weathering, leaching of rocks and soil, domestic and municipal effluents (Sarath Prasanth et al. 2012). The SO_4^{2-} values varied from 25-180 mg/l and 13-221 mg/l in pre and post-monsoon seasons. The leachable sulphate presents in fertilizer and other human influences increase the sulphate concentration in groundwater due to breaking of sulphates (Craig and Anderson, 1979). The NO_3^- values varied from 6-166 mg/l and 1-288 mg/l in pre and post-monsoon seasons. The high Nitrate values are indicating leaching of organic substances from weathered soil. The seasonal variations of nitrate were caused due to change in the volume of drainage reaching the water table and not by the change in concentration. The Ca^{2+} values ranges from 24-188 mg/l and 20-186 mg/l and Mg^{2+} values varying from 10-138 mg/l and 8-160 mg/l in pre and post-monsoon seasons, respectively. The TH values range from 101-1038 mg/l and 97-1023 mg/l in pre and post-monsoon seasons. The hardness of the water is due to the presence of alkaline earths such as calcium and magnesium. The Na^+ values range from 4 –530 mg/L and 30 – 477 mg/L and K^+ values range from 2-130 mg/l and 1-129 mg/l in pre and post-monsoon seasons, respectively. The fluoride concentration varied from 0.52-3.40 mg/l and 0.08- 2.60 mg/l in pre and post-monsoon seasons. The maximum tolerance limit (1.5 mg/l) recommended by World Health Organization (WHO 2011). Ingestion of water with fluoride concentration above >1.5 mg/l causes fluorosis (Madhnure et al. 2007).

Due to highly weathered formation and long-time pumping for irrigation use are the responsible factors for the leaching of fluoride minerals from their parent rocks. Further concentration has been brought about due to semi-arid climate of the region and long residence time of groundwater in the aquifer (Wodeyar and Sreenivasan 1996). The influence of local lithology and soil aided by other factors like very low freshwater exchange due to the semi arid climate of the regions (average daily temperature 30^0C and average rainfall of 638mm) is responsible for higher concentration of fluoride in the groundwater of the region. Higher concentration was noted that due to presence of dominant fluoride bearing minerals like apatite [$Ca_5(PO_4)_3$ (F, Cl, OH)], hornblende [$Ca_2(Mg, Fe, Al)_5$ $(Al, Si)_8$ O_{22} $(OH)_2$], and biotite [$K(Mg, Fe)_3$ $(AlSi_3O_{10})$ $(F, OH)_2$] which have enhanced the fluoride concentration. Higher concentration of fluoride was noted in granites due to the easy accessibility of fluoride minerals due to the higher degree of weathered and fractured zones in granites.

Hydrochemical facies

Dissolution and deposition of fluoride is controlled by chemical type of groundwater. Hence, major ion chemistry of groundwater is examined by using (Piper, 1994) trilinear diagram to identify chemical alteration in groundwater. The Piper diagram consists of two lower triangular fields and a central diamond-shaped field. All the three fields have incorporation of major ions only. The triangular fields are plotted separately with epm values of cations (Ca+Mg) alkaline earth, (Na+K) alkali, HCO_3 weak acid, and (SO_4+Cl) strong acid. Water facies can be identified by projection of plots in the central diamond-shaped field as per the classifications made by (Karanth, 1987). Aquachem 4.0 scientific software is used for the plotting of piper trilinear diagram. The piper diagram is dominated in pre and post-monsoon seasons are CaHCO3, mixed CaNaHCO3, NaCl and mixed CaMgCl water type (**Fig. 3**).

Gibbs diagram

Gibbs diagram is widely used to establish the relationship of water composition and aquifer lithological characteristics. Three distinct fields such as precipitation dominance, rock dominance and evaporation dominance areas are shown in the Gibbs diagram (Gibbs, 1970). MS-Excel 2007 version is used for the plotting of Gibbs diagram. The predominant samples fall in the rock dominance and evaporation dominance field of the Gibbs diagram in the both pre and post-monsoon seasons **(Fig. 4)**. The rock dominance field indicates the interaction between rock chemistry and the chemistry of the percolated waters under the subsurface. The Evaporation results in increased TDS relative to the confined water with relation to high ratios of dominant cations and anions, especially sodium ions, due to $CaCO_3$ precipitate caused by the combination of Ca^{2+} and HCO_3^-. Huge quantities of irrigation return flow elevated groundwater level, hence increases evaporation and induce salinization.

Spatial distribution

Arc GIS 10.1 version used to the spatial distribution maps of the study area. The spatial distribution of fluoride in the groundwater **(Fig. 5)** was attempted to identify regions and locations of widespread fluorosis. During PRM, fluoride (0.52-1.5 mg/l) was noted in 67% of the samples in fourteen locations are within in the permissible limits and higher concentration (>1.5 mg/l) was observed in 33% of samples in seven locations are exceed the permissible limit (WHO, 2011). In general, fluoride was higher in both litho units. During POM, fluoride (0.08-1.5 mg/L) was noted in 71% of the samples in fifteen locations are permissible limit. Higher concentration (>1.5 mg/L) was observed in 29% of samples in six locations are exceed the permissible limit (WHO 2011). High concentration of fluoride is observed in Korukondapally village in pre and post-monsoon seasons. In general, seasonal fluctuations were noted higher when compared with litho logical influence during both the seasons.

PRINCIPAL COMPONENT ANALYSIS (PCA)

The principal component analysis obtained by Initial Eigen values and percent (%) of the variance on the groundwater chemical data and reduced the dimensionality from the 14 original physico-chemical parameters to four principal components (PCs) (**Table 3**) for pre and post-monsoon seasons by using Statistical Package for Social Sciences (SPSS) software.

PCA was applied to the varimax normalized data to compare the compositional patterns between the analyzed water samples and to identify the factors that influence each one. The factor loading are classified (Mor et al., 2006) as strong, moderate and weak corresponding to absolute loading values of >0.75, 0.75-0.50 and 0.50-0.30, respectively. The output of the final rotated loading matrix obtained from the present data indicates in the pre-monsoon season obtained the results that the three principal component analysis explains 80.90% of the total variance, PC1 represent 43.56% of variance with loading of SO4, Mg, Ca, Cond., TDS, CL, NO3, Na and HCO3 due to cations and anions resulted from the mineral weathering and water-rock interactions in the aquifer (Purushotham et al., 2011). PC2 is represent 27.79% of the total variance and had high loadings on Cond., TDS, CO3, Na and F which indicates ion-exchange and carbonate weathering and leaching from fluoride-bearing minerals (apatite, hornblende, mica, etc.). PC3 is represent 9.55% of the total variance and high loadings on K^- which indicates weathering reactions involving silicate minerals, possible K-feldspar and positive loading for nitrate is pre-dominant,

In the Post-monsoon season obtained the results of the three factor analysis's explains 85.33% of the total variance, the four PC1, 2, 3 and 4 were found to be responsible for the variations in groundwater quality explains 50.23%, 24.29% and 10.80 of total variance respectively in the data. Here again PC1 show high loadings on SO4, Mg, Ca, Cond, TDS, Cl, NO3 and Na similar to the PRM PC1. The PC2 is influenced by pH, CO3, Na, HCO3 and F, which indicating pre-dominant of K-feldspar, salinity of the water and high loading of nitrate in areas huge amount of fertilizers being used. PC3 includes variable like K negative loading which indicate the leaching from irrigation of agricultural field.

In summary, it seems that different hydro-geochemical processes like weathering, ion-exchange, and anthropogenic inputs are the key factors that dominate the groundwater chemistry in the study area. At most, principal component analyses substantiate the findings of previous sections and provide greater confidence in data interpretation.

CONCLUSION

The present study area in and around of Kesamudram mandal, Mahabubbad district, Telangana State, India and total spread of 148 km^2 area. It is entirely underlined by Peninsular gneissic complexes. The geology of the study area is grey and pink granites occupy dominant portion of the study area. These rocks are composed of quartz, feldspar, biotite and hornblende. The present study is an attempt to demarcate fluoride-vulnerable zones and to identify major geochemical process controlling the incidence of fluoride in the groundwater of study area. The fluoride concentration ranges from 0.52 to 3.40 mg/l and 0.08 to 2.60 mg/l during pre and post-monsoon seasons, respectively. Results showed that collected water samples were contaminated by presence of fluoride ion. During PRM 33% and POM 29% of samples recorded higher fluoride when compared with World Health Organization tolerance limit (1.5 mg/l). According to the Gibbs diagram majority of samples are fall in the rock and evaporation dominance field in the both pre and post-monsoon seasons. The four PCs were extracted during PRM and POM indicating the combined action of hydro geochemical processes like weathering, ion exchange, and anthropogenic inputs are the key factors that determine the groundwater chemistry in the study area.

ACKNOWLEDGEMENT

We are thankful to Head, Department of the Applied Geochemistry, Osmania University, Hyderabad, for providing necessary laboratory facilities. The authors gratefully acknowledge the UGC, New Delhi for providing Research Fellowship in Science for Meritorious Scholars (RFSMS) during the progress of the research work.

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Table 1 Year wise rain fall distribution in the study area (in mm)

Year	**Annual rain fall**
2010-11	632.60
2011-12	754.40
2012-13	1150.20
2013-14	1158.40
2014-15	753.00
2015-16	930.20
2016-17	1635.20
2017-18	991.80
2018-19	839.00
2019-20	1144.00
Minimum	**632.60**
Maximum	**1635.20**
Average	**998.88**

Table 2 Statistical summary of physical and chemical parameters of groundwater samples in pre and post-monsoon seasons

Parameter	Pre-monsoon season					Post-monsoon season (August)				WHO, 2011
	Min	Max	Ave	% of samples exceeded the limits		Min	Max	Ave	% of samples exceeded the limits	
pH	7.36	8.56	8.14	10		6.76	8.0	7.61	0	**6.5 - 8.5**
EC (µS/cm)	320	3680	1607	57		288	3230	1205	29	**1500**
TDS (mg/l)	205	2355	1028	91		153	1730	643	62	**500**
Ca^{2+} (mg/l)	24	188	91	52		20	186	79	48	**75**
Mg^{2+} (mg/l)	10	138	51	38		8	160	48	24	**50**
TH as CaCO3	101	1038	434	33		97	1022.8	396	19	**500**
Na^{+} (mg/l)	45	530	187	24		30	477	155	29	**200**
K^{+} (mg/l)	2	130	11	5		1	129	9	5	**12**
HCO_3^- (mg/l)	142	640	420	33		130	670	371	5	**500**
CO_3^- (mg/l)	0	66	26	-		0	60	24	-	**30**
Cl^- (mg/l)	12	680	172	19		5	678	138	14	**250**
NO_3^- (mg/l)	6	166	51	38		1	288	56	43	**45**
SO_4^{2-} (mg/l)	25	180	62	-		13	221	59	-	**250**
F^- (mg/l)	0.52	3.40	1.45	33		0.08	2.60	1.18	29	**1.5**
Gibbs I (meq/l)	0.35	0.89	0.65	-		0.29	0.83	0.61	-	**-**
Gibbs II (meq/l)	0.11	0.65	0.36	-		0.06	0.71	0.32	-	**-**

*Min-Minimum, Max-Maximum and Mean

Table 3 Principal component analysis for water samples during pre and post-monsoon seasons

Parameters	Pre-monsoon			Post-monsoon		
	PC1	PC2	PC3	PC1	PC2	PC3
SO_4^{2-}	**.926**	.059	.061	**0.962**	-0.038	0.038
Mg^{2+}	**.924**	.198	.115	**0.824**	0.352	0.078
Ca^{2+}	**.914**	.047	.003	**0.82**	0.109	0.146
Cond	**.816**	**.563**	.053	**0.907**	0.403	0.013
TDS	**.816**	**.563**	.053	**0.908**	0.402	0.011
Cl^-	**.812**	.410	.008	**0.932**	0.222	-0.09
NO_3^-	**.565**	.119	-.350	**0.911**	0.222	-0.09
pH	.077	**.842**	.006	0.114	**0.893**	-0.023
CO_3^{2-}	.336	**.789**	.057	0.314	**0.802**	-0.08
Na^+	**.585**	**.721**	-.120	**0.761**	**0.499**	-0.24
HCO_3^-	**.568**	**.691**	.141	0.464	**0.761**	0.13
F^-	-.139	**.649**	-.484	-0.042	**0.584**	**-0.692**
K^+	.024	.048	**.908**	-0.042	0.11	0.89
Initial Eigen	**5.66**	**3.61**	**1.24**	**6.53**	**3.16**	**1.40**
% of Variance	**43.56**	**27.79**	**9.55**	**50.23**	**24.29**	**10.80**
Cumulative %	**43.56**	**71.35**	**80.90**	**50.23**	**74.52**	**85.33**

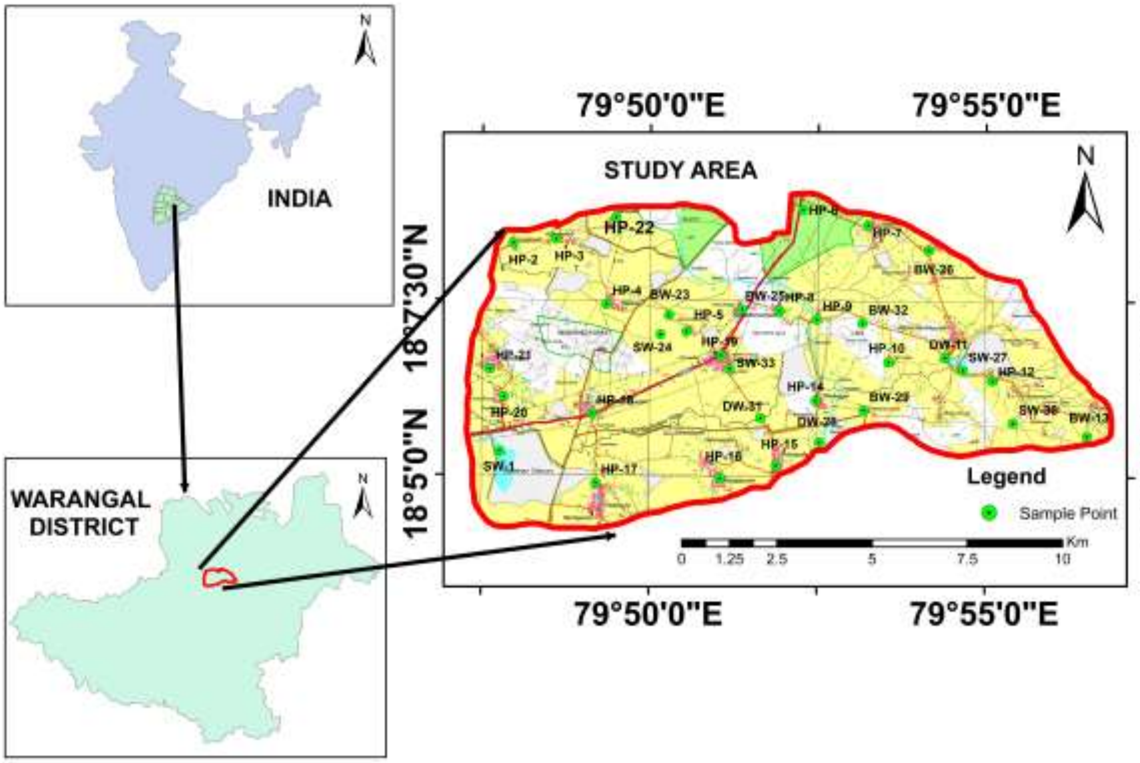

Fig. 1: Study area with water sample locations

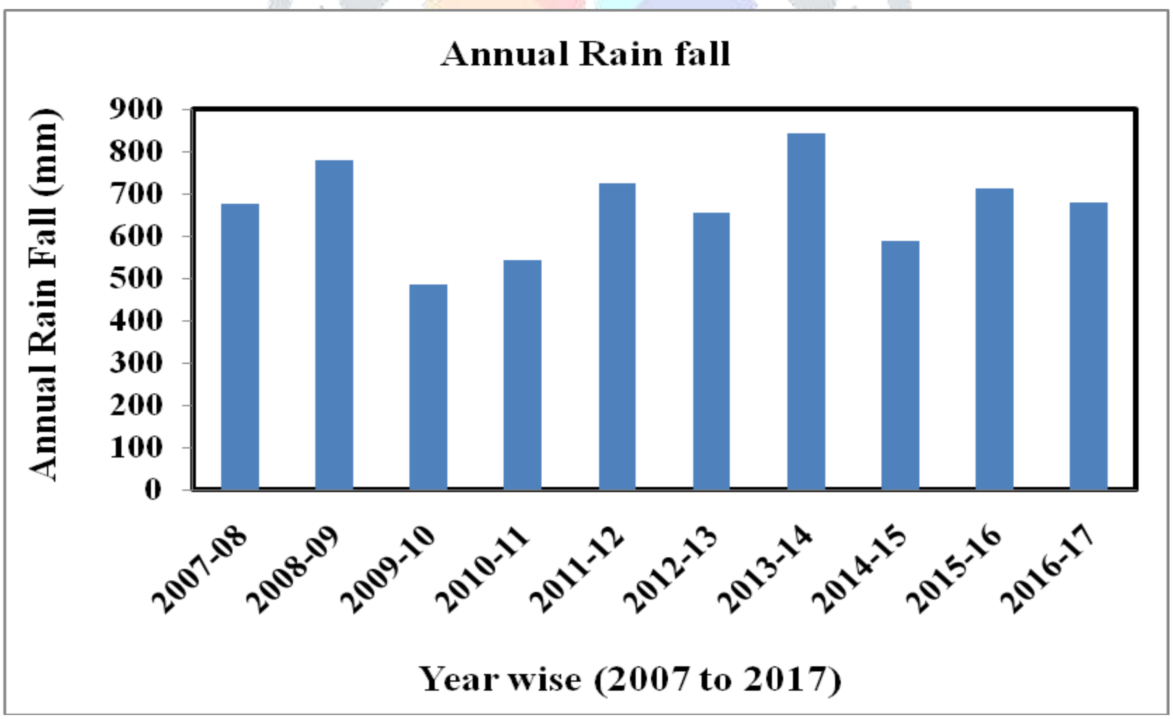

Fig. 2: Year wise rain fall distribution in the study area (in mm)

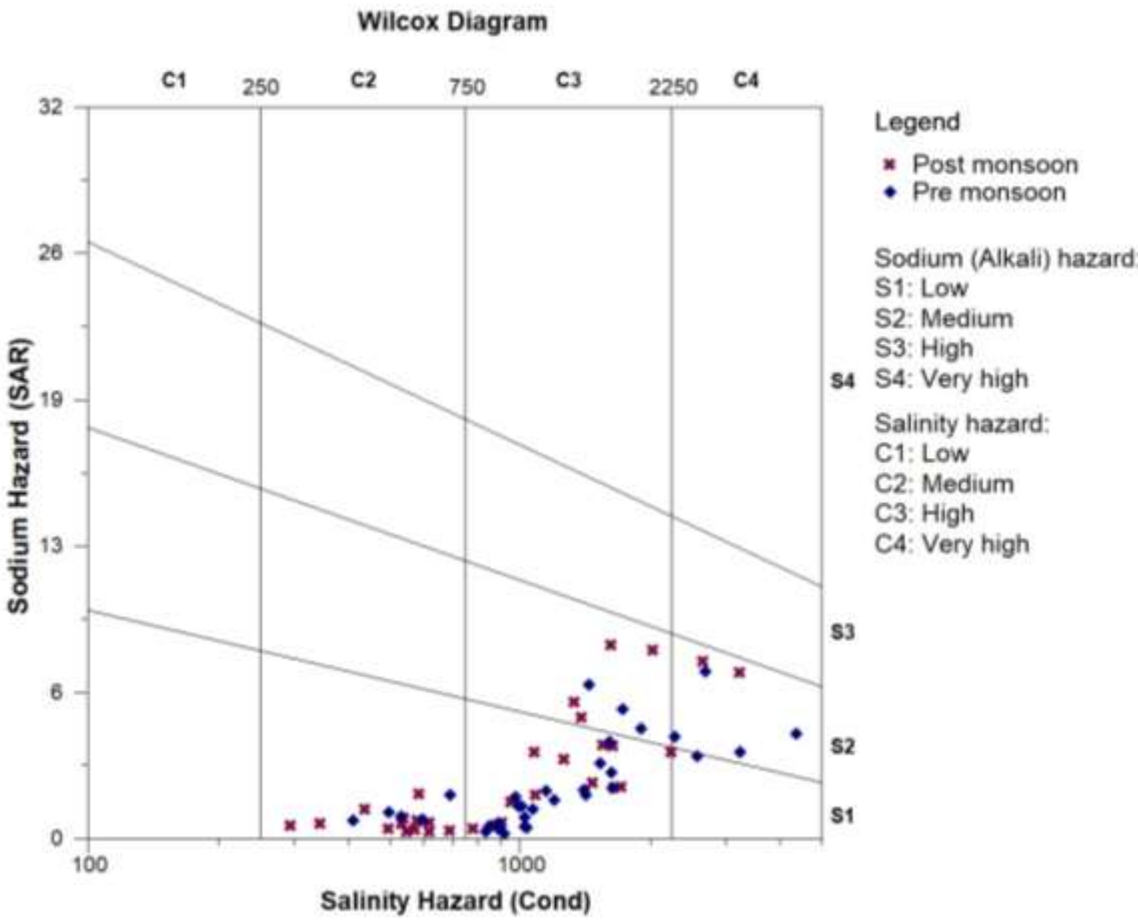

Fig. 3: USSL classification of groundwater in pre-monsoon and post-monsoon season

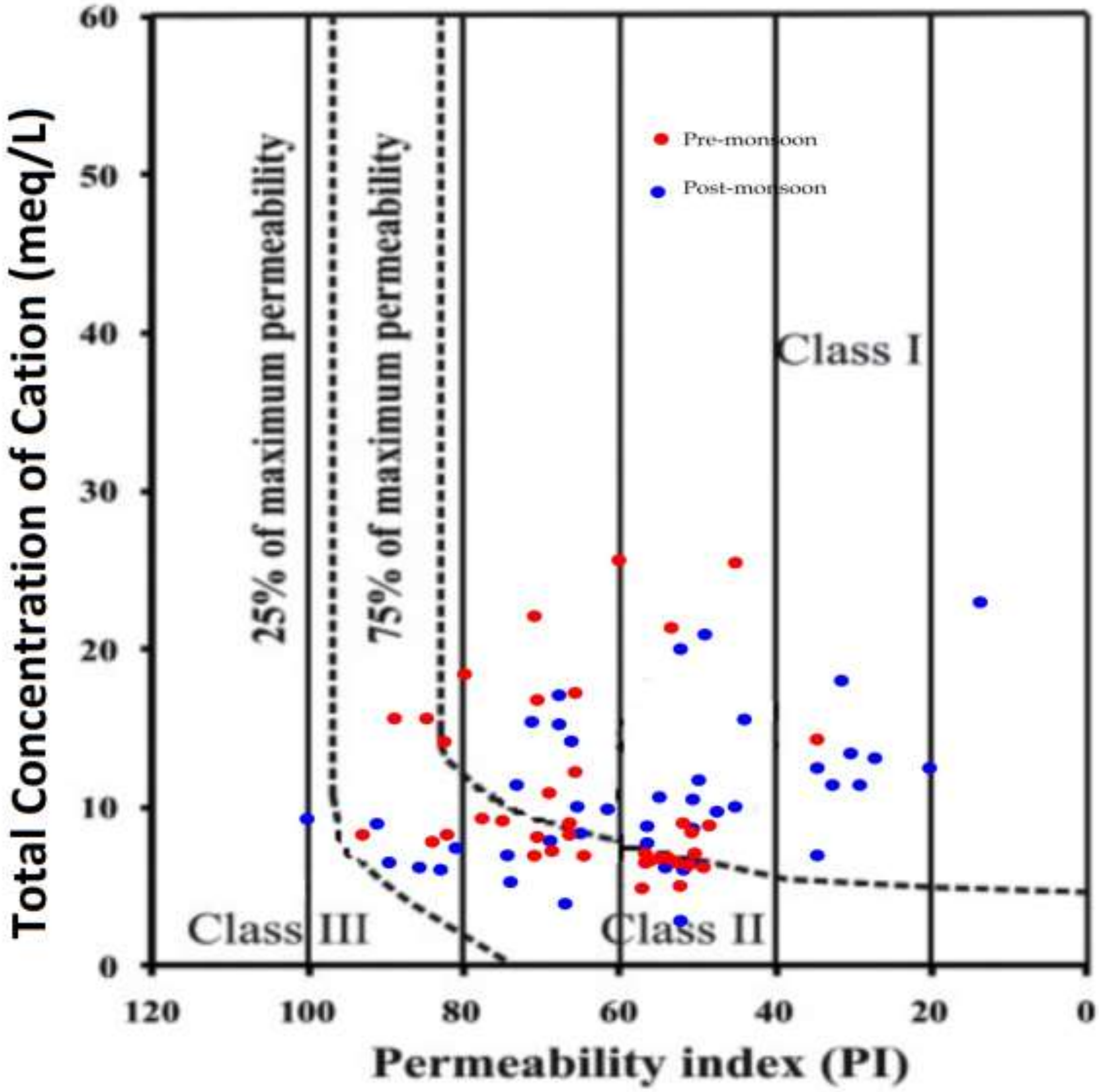

Fig. 4: Doneen Classification (1964) of Irrigation Water based on the Permeability index in pre and post-monsoon seasons

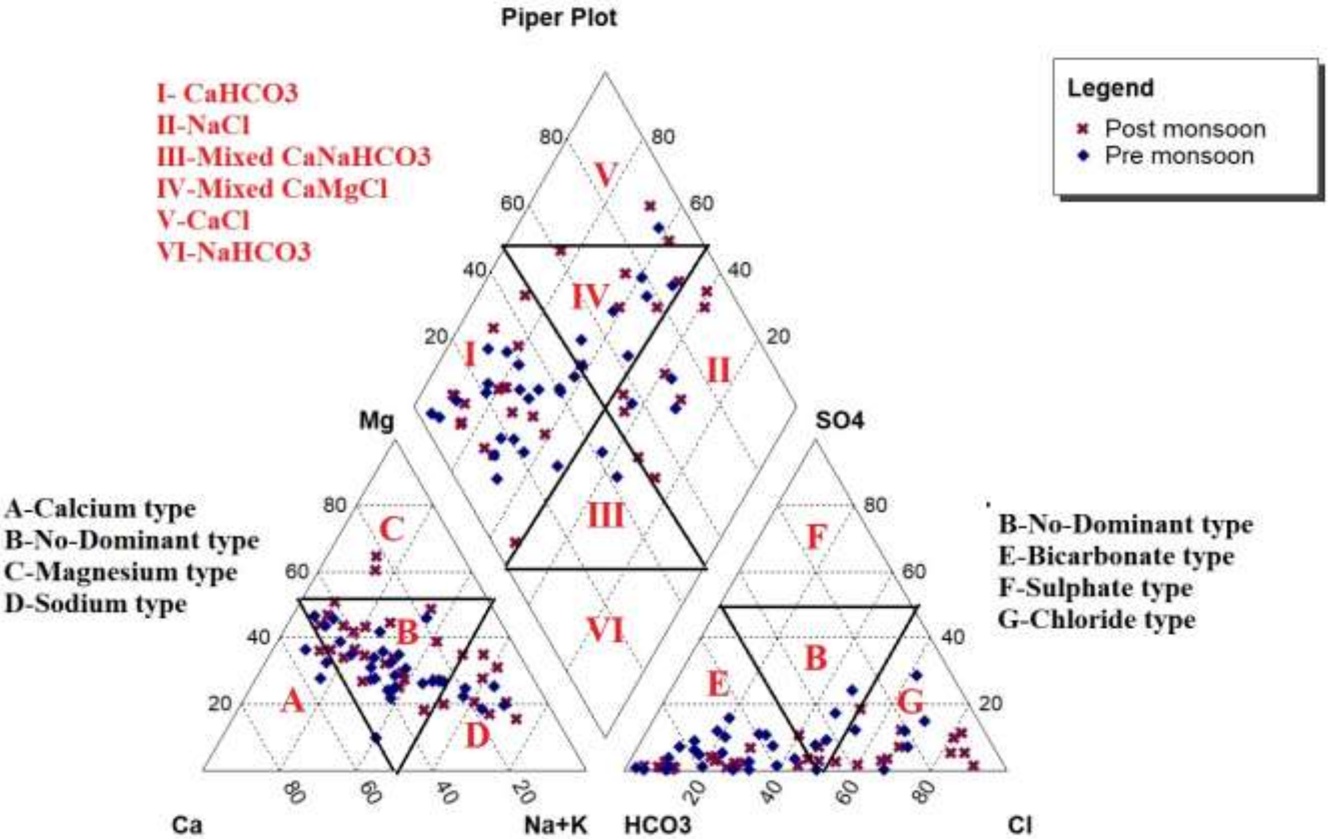

Fig. 5: Hydrochemical facies classification of groundwater from the study area in pre and post-monsoon seasons

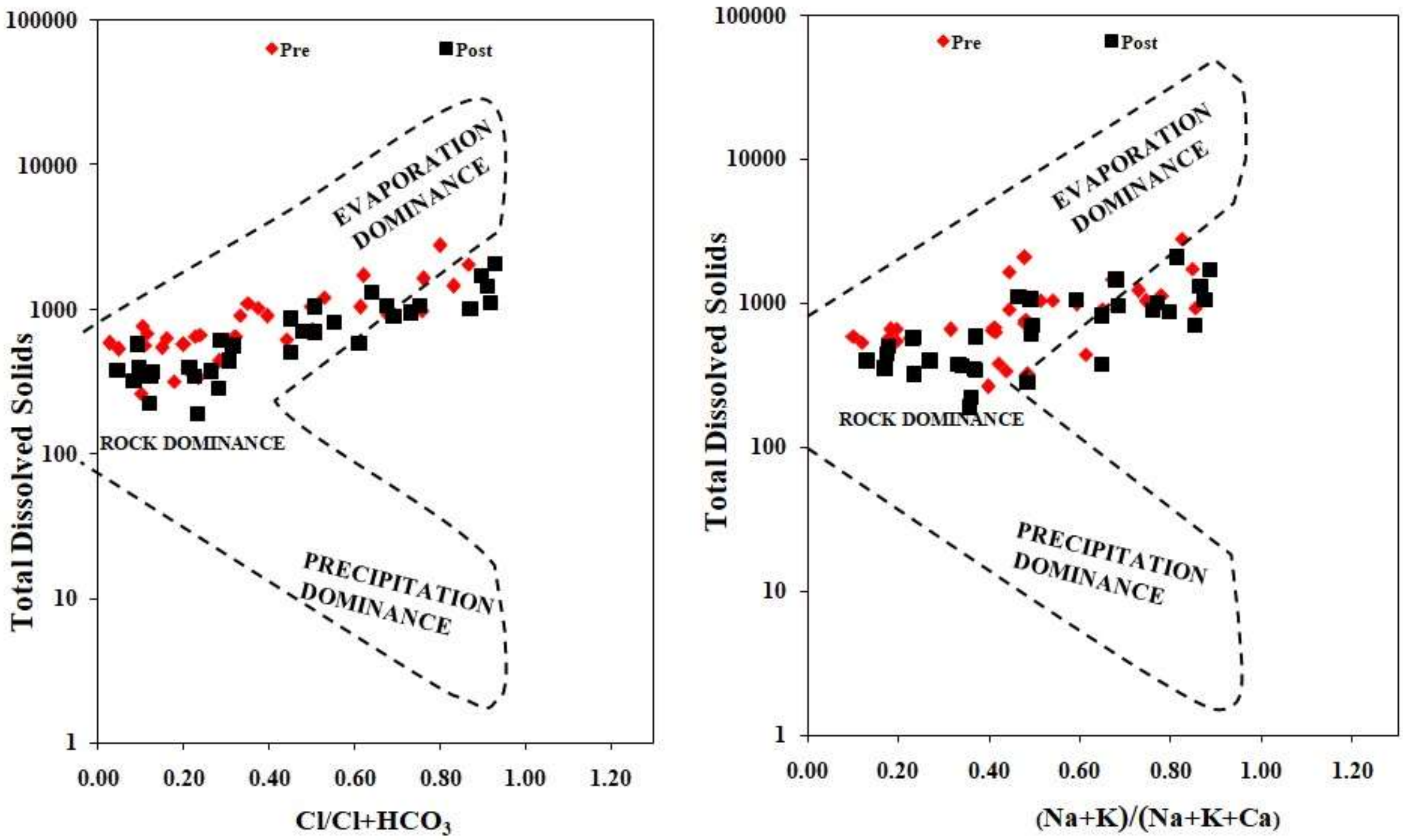

Fig. 6: Gibb's plot for the groundwater from the study area in pre and post-monsoon seasons