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Spatial Distribution of Nitrate and Fluoride Contaminations in Groundwater, Shaligowraram Mandal, Telangana State, India

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ABSTRACT

The elevated levels of groundwater contamination in the shallow, hard-rock aquifer of Shaligowraram Mandal, located in the semi-arid region of Telangana State, South India, are primarily influenced by hydrogeochemical regulatory factors. A total of 42 groundwater samples were systematically collected and analyzed during both the pre-monsoon and post-monsoon seasons to assess spatial and seasonal variations in water quality. The evaluation focused on hydrochemical facies, water-rock interaction processes, and spatial distribution patterns. Notably high concentrations of nitrate (NO_3^-) and fluoride (F^-) were detected, particularly in the central, north-eastern, north-western, eastern, and south-eastern parts of the study area. During the pre- and post-monsoon periods, nitrate exceeded the permissible limit of 45 mg/L in approximately 88% and 63% of the samples, respectively, while fluoride surpassed the acceptable threshold of 1.5 mg/L in about 45% and 32% of the samples. Gibbs diagrams indicate that the dominant geochemical processes influencing groundwater chemistry in the region are rock-water interaction and evaporation, reflecting the arid climatic conditions and lithological characteristics of the area.

Keywords: Nitrate, Fluoride, Hydrochemical facies and Gibb's plot

Introduction

Natural resources are extremely important to us. In particular, clean water and fresh air are necessary for human survival on this planet. The two most important sources of water for industrial, agricultural, and domestic uses are surface water and groundwater (Laxman et al. 2019; Muralidhara and Sunitha 2020). A third of the world's population, especially in arid and semi-arid nations, depends on groundwater for their daily needs, especially for drinking, because there is a shortage of surface water (USEPA 2014). Groundwater quality is steadily declining as a result of both natural environmental changes and human activities like farming and industry. This has led to a crisis of water scarcity and a number of other environmental issues (Satyanarayana et al. 2017; Ali et al. 2021). Poor groundwater quality has an impact on worldwide nitrate and fluoride levels. In recent years, a number of research on fluoride and nitrate levels in water have been undertaken in the world's arid and semi-arid regions.

Nitrate (NO_3^-) concentrations are commonly observed in nitrogen-based fertilizers. Improper disposal of waste from animal feedlots, dairies, agricultural land, and landfill leachate can increase the nitrate concentration of aquatic bodies (Ali et al. 2021). The presence of NO_3^- in drinking water poses a risk to animal and human health. The drinking water standards are intended to prevent bad health consequences in neonates, such as methemoglobinemia. According to Eggers et al. (2018), pregnant women, babies, and children are especially sensitive to high NO_3^- concentrations. Increased NO_3^- levels in water bodies can lead to poor water quality, eutrophication, and harmful algae blooms (Briki et al. 2017). Excessive NO_3^- concentrations in drinking water can cause diseases such as spontaneous miscarriages, thyroid disorders, cancer, teratogenesis, mutagenesis, and methemoglobinemia (blue baby syndrome) (Wu et al. 2018).

likewise, fluorine is a common element found in soil and water that was identified by the USEPA in 2014 as a dangerous chemical contaminant (Amiri and Berndtsson 2020; Ali et al. 2021; Sohrabi et al. 2021). Fluoride is naturally present in granite, gneisses, basalt, and shale rocks. Fluoride ions enter aquatic environments through a variety of pathways, including beryllium extraction, brick and iron works, aluminum smelters, coal-fired power plants, and electroplating enterprises. Fluoride in drinking water at certain amounts can lead to tooth decay (<0.50 mg/L), fluorosis (1.5-5 mg/L), and skeletal fluorosis (5-40 mg/L) (Kimambo et al. 2019; Vithanage and Bhattacharya 2015). High F^- concentrations (>10 mg/L) have been linked to several health issues, including arthritis, neurological diseases, thyroid, cancer, infertility, and hypertension. Fluoride also affects the teeth and skeleton, causing changes in DNA structure (Kimambo et al. 2019).

The study region and general Geology

The Shaligowraram region extends from latitude 17.2308 to 17.3522 north and longitude 79.3036 to 79.4706 east (Fig. 1). The total area covered is 220 km². The research area is made up of rugged terrain and monolithic boulders on the boundaries of the region. The research area is semi-arid and dominated by granitic topography (Fig. 2). Groundwater samples were collected in one-litre bottles, cleaned with 1:1 dilute nitric acid, and rinsed with distilled water. Before collecting the samples, they were washed again with the same sample, as per standard technique (APHA 2012). After collecting water samples, pH/EC was measured using hand-held meters (Hanna USA H-198130). The TDS values are determined by multiplying 0.64 by the EC values.

Ion levels such as calcium, magnesium, sodium, and potassium as cations, and bicarbonate, chloride, sulfate, nitrate, and fluoride as anions were analyzed following standard protocols. Key parameters were determined using specific methods: total hardness and calcium via titration, sodium and potassium through flame photometry, sulfates and nitrates using a spectrophotometer, and fluoride via an ion-specific electrode. Analytical accuracy was confirmed by considering the total levels of cations and anions, which revealed a $\pm 5\%$ margin of error, underscoring the reliability of the data.

Results and discussion

General characteristic of groundwater chemistry

The study region's groundwater chemistry was studied in accordance with standard drinking water criteria (BIS 2012). The pH values in the study area range from 7.80 to 9.40 with an average of 8.26, and 7.56 to 9.22 with an average of 8.28 in the pre- and post-monsoon seasons, respectively (Table 1), indicating basic/alkaline conditions. The study region's drinking water falls within the allowed pH range of 6.5 to 8.5 (BIS 2012). In the pre-monsoon season, the average EC was 1623 $\mu\text{S}/\text{cm}$, but in the post-monsoon season it was 1197 $\mu\text{S}/\text{cm}$ (see Table 1).

Spatial distribution of Nitrate

The cumulative trend of groundwater nitrate contamination over the last few decades has been driven by fast urban expansion, industrial development, and the widespread use of nitrate-based fertilizers for agricultural and horticultural applications. Excessive nitrate in drinking water can be harmful to human health (Zhang et al. 2018; Wagh et al. 2019).

In both seasons, NO_3^- levels range from 18 to 188 mg/L, with a mean of 53 mg/L, and 12 to 140 mg/L, with a mean of 50 mg/L in the pre- and post-monsoon seasons (Table 1 and Figure 3). The drinking water criterion for nitrate is 45 mg/L (WHO 2011). It has no health risk to people; 45-100 mg/L causes health impacts in children and adults; and >100 mg/L poses a very high health risk. A considerable increase in the percentage of samples classified as "No health risk" from 50% in the pre-monsoon season to 67% in the post-monsoon season, indicating a diluting effect caused by monsoon rains. The percentage of groundwater samples in the "High health risk" category decreases significantly from 45% pre-monsoon to 26% post-monsoon. The "Very high health risk" class has slightly increased from 5% pre-monsoon to 7% post-monsoon, demonstrating continuing localized nitrate contamination in the research area (Fig. 4).

Oenema et al. (2005) discovered that nitrate mostly percolates into the aquifer regime from vast farming, horticulture, residential, and manufacturing wastes. Fertilizers including diammonium phosphate, urea, and superphosphate are widely used and can seep into the subsurface. Because nitrate fertilizers are widely used, nitrate can drain away from soils, increasing nitrate levels in groundwater (Jalali 2011; Lokesh, 2013; Amiri et al. 2021).

Spatial distribution of Fluoride

Notwithstanding a regulatory maximum fluoride concentration in drinking water of 1.5 mg/L (BIS 2012), excessive exposure is associated with dental and skeletal fluorosis (Laxman et al. 2019). Spatial fluoride interpolation within the observed geographical area exhibits considerable variance, ranging from 0.24 to 2.70 mg/L with a mean of 1.30 mg/L, and 0.15 to 2.11 mg/L with a mean of 1.08 mg/L during both monsoons. Notably, the recommended limit of 1.0 mg/L (BIS 2012) has surpassed in approximately 67% and 64% of samples, respectively (Table 1 and Fig. 5). High fluoride spatial interpolation is characterized by a distinctive pattern, as illustrated in Fig. 6. Fluoride distribution is predominantly observed in the northern, northwest, northeastern, western, eastern and southwestern regions, with the alkaline nature of the water contributing to anionic exchange and increased fluoride levels in the aquifer regime (Saxena and Ahmed 2003).

Hydrochemical facies

A Piper diagram reveals that two triangular fields comprise the cation and anion areas at the base, while a central diamond-shaped field represents the Piper Trilinear classification of a given study region (Piper, 1944). The region's hydrochemical facies are categorized into nine groups during the pre and post-monsoon seasons, with a majority of samples demonstrating strong acid predominance over weak acid, alkaline earth exceeding alkalis, and a mixed facies type, characterized by negligible cation-anion exceedance.

Gibb's diagram

A Gibbs diagram was utilized for evaluating the impact of rock degradation, precipitation, and evaporation on the chemical composition of groundwater (Gibbs 1970). The modified Gibbs plot was employed to elucidate the prevalent geochemical processes in groundwater (Marandi and Shand 2018). Groundwater chemistry is influenced by a myriad of processes and factors, including mineral solubility and availability, exchange processes, geochemical environment, groundwater movement, residence time, and regional climatic conditions. Gibbs' ratio-I and ratio-II values ranged from 0.45 to 0.91 meq/L and 0.48 to 0.88 meq/L, with a mean of 0.69 meq/L, and from 0.34 to 0.79 meq/L and 0.34 to 0.76 meq/L, respectively (Table 1). Figure 8 indicates that the majority of groundwater samples are situated within the rock-dominant zone for both pre-monsoon and post-monsoon seasons (Amiri et al 2020). This demonstrates that rock degradation, particularly ion exchange, exerts a significant influence on groundwater chemistry, with the granitic gneiss terrain contributing to the widespread weathering of rock-forming minerals. However, a few regional anomalies suggest the presence of anthropogenic impacts, as evidenced by increased concentrations of sodium, chloride, and total dissolved solids (Saxena and Ahmed 2001; Satyanarayana et al. 2017).

Conclusions

The pH levels within the study area exhibit a range of 7.80 to 9.40 during the pre-monsoon season and 7.56 to 9.22 during the post-monsoon season, characterised by an alkaline nature. Geochemical analysis indicates that the highest fluoride concentrations occur in the northern, northwest, northeastern, western, eastern, and southwestern regions, where granitic terrain is prevalent and rich in fluoride-bearing minerals. Elevated nitrate concentrations suggest potential contamination from residential sewage and fertilisers penetrating the subsurface. Piper diagram analysis indicates that both seasons display rock dissolution patterns, with predominant ion concentrations of Na-Ca-Mg-Cl-HCO₃ and Mixed Na-Ca-Mg-Cl, respectively. Gibbs diagram results reveal a predominantly rock-dominated regime, with minimal evaporation influence, whereas groundwater quality variation is largely ascribable to in situ rock-water interactions, influenced by anthropogenic activities and ion exchange processes within the aquifer.

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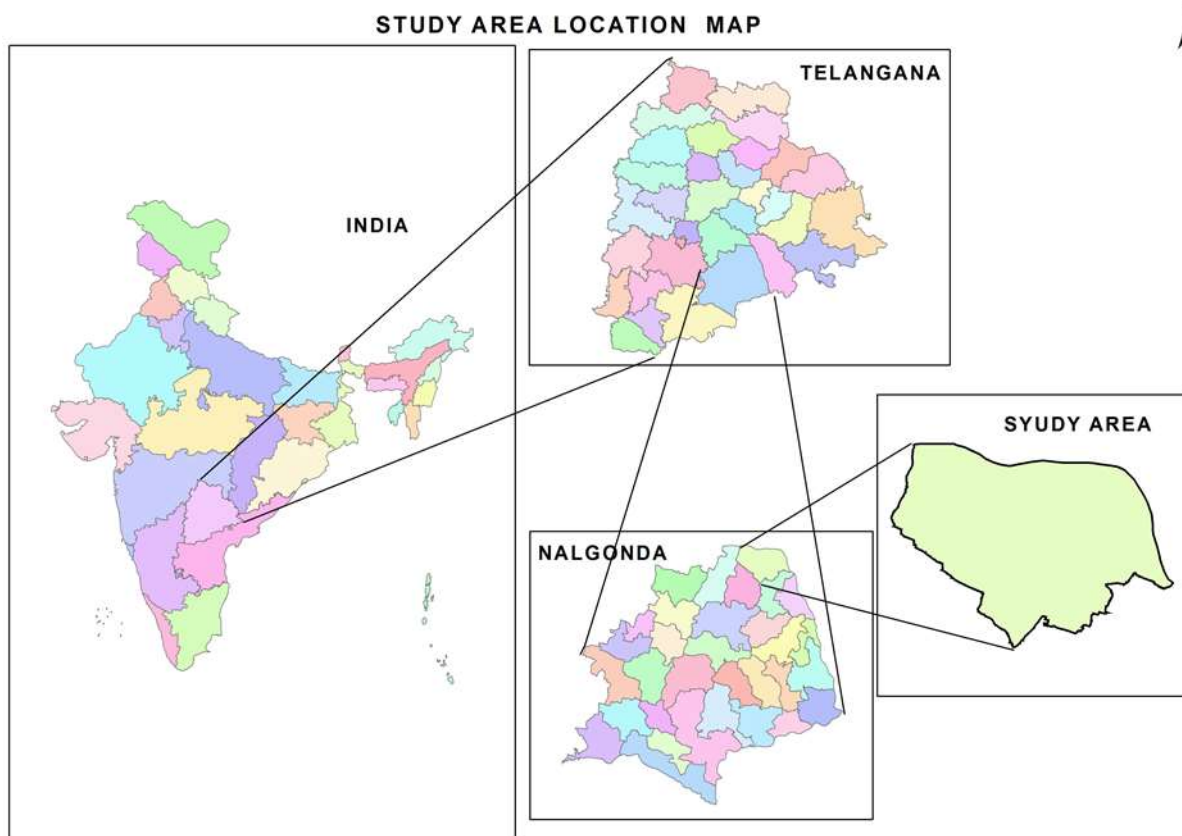


Fig. 1 Location map of the study area

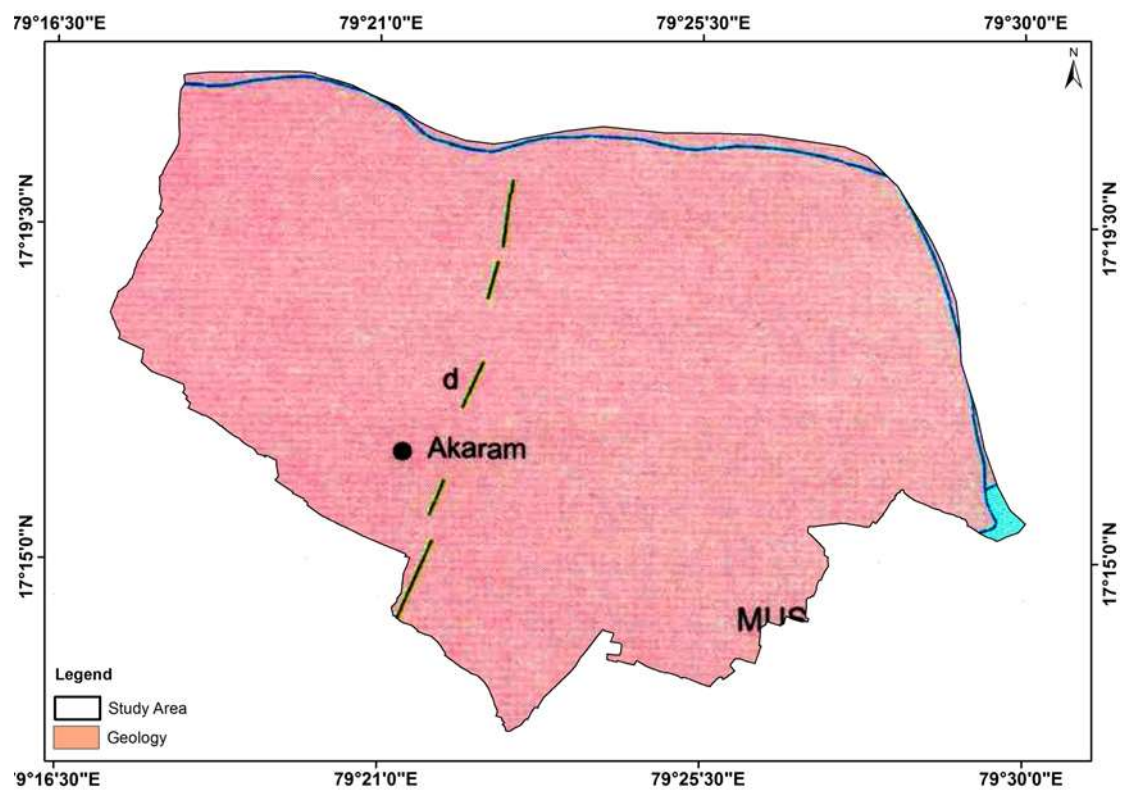


Fig. 2 Geology map of the study area

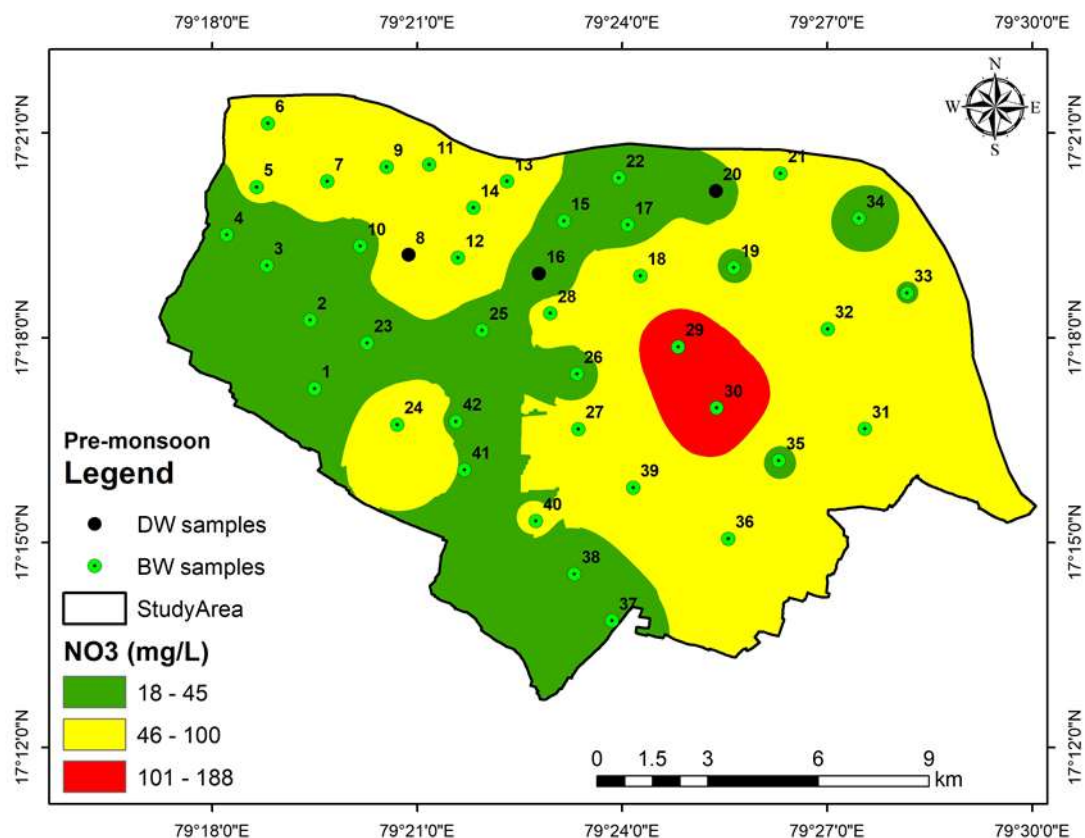


Fig. 3 Spatial distribution map of Nitrate concentration in pre and post-monsoon seasons

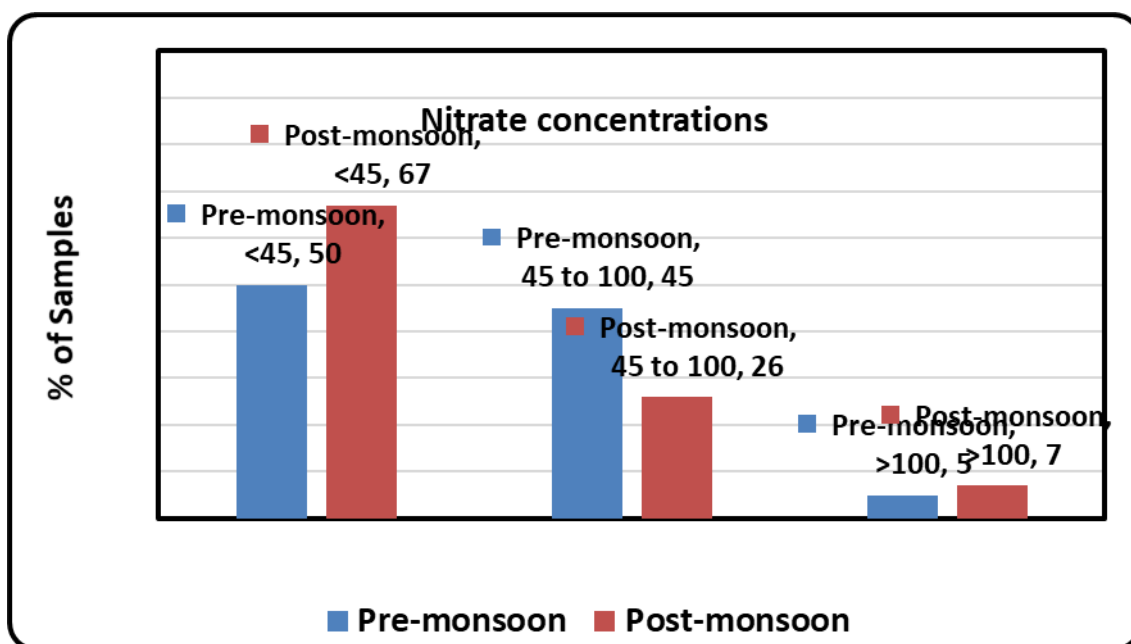


Fig. 4 Nitrate concentration levels in pre and post-monsoon seasons

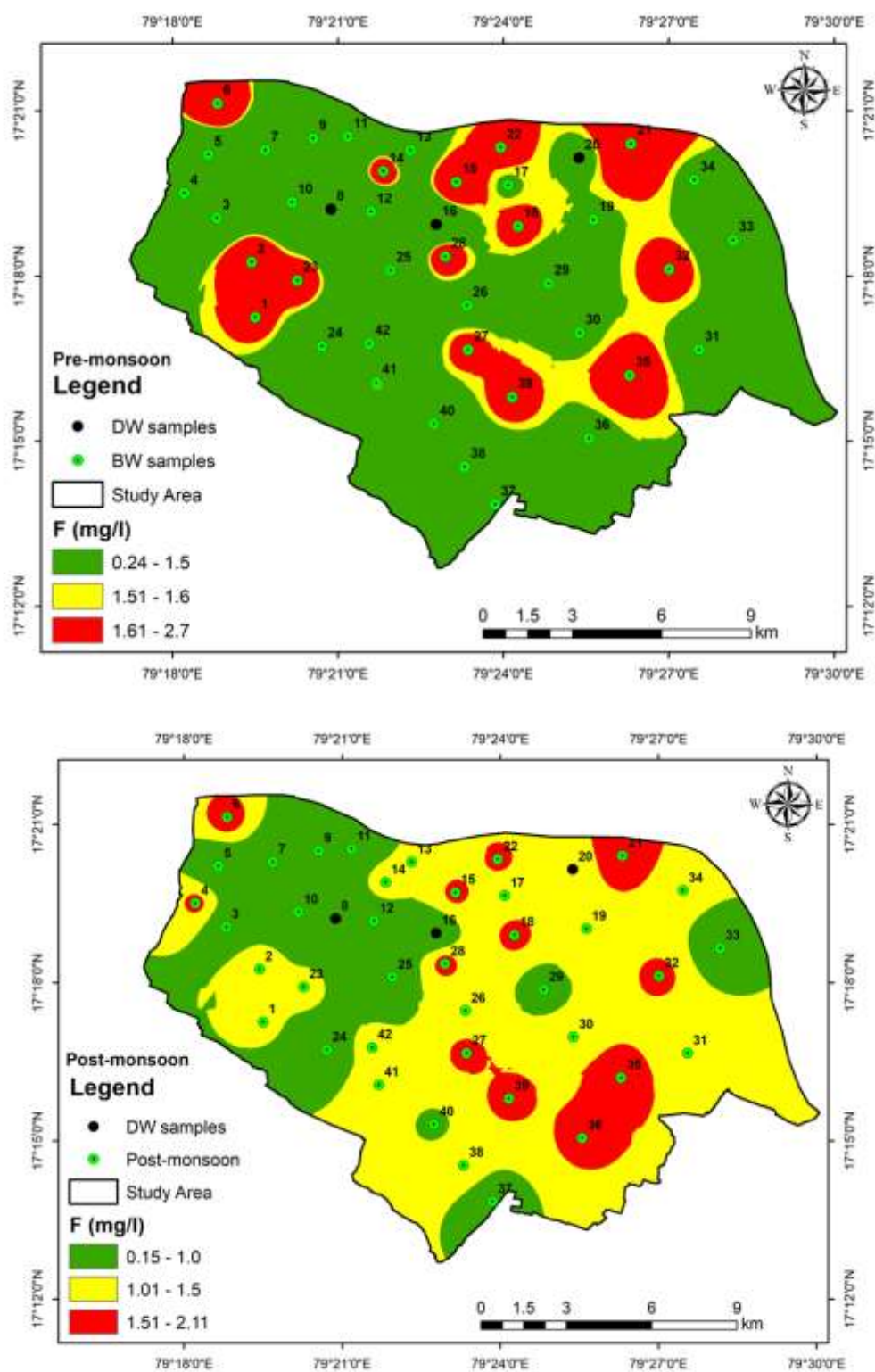


Fig. 5 Spatial distribution map of Fluoride concentration in pre and post-monsoon seasons

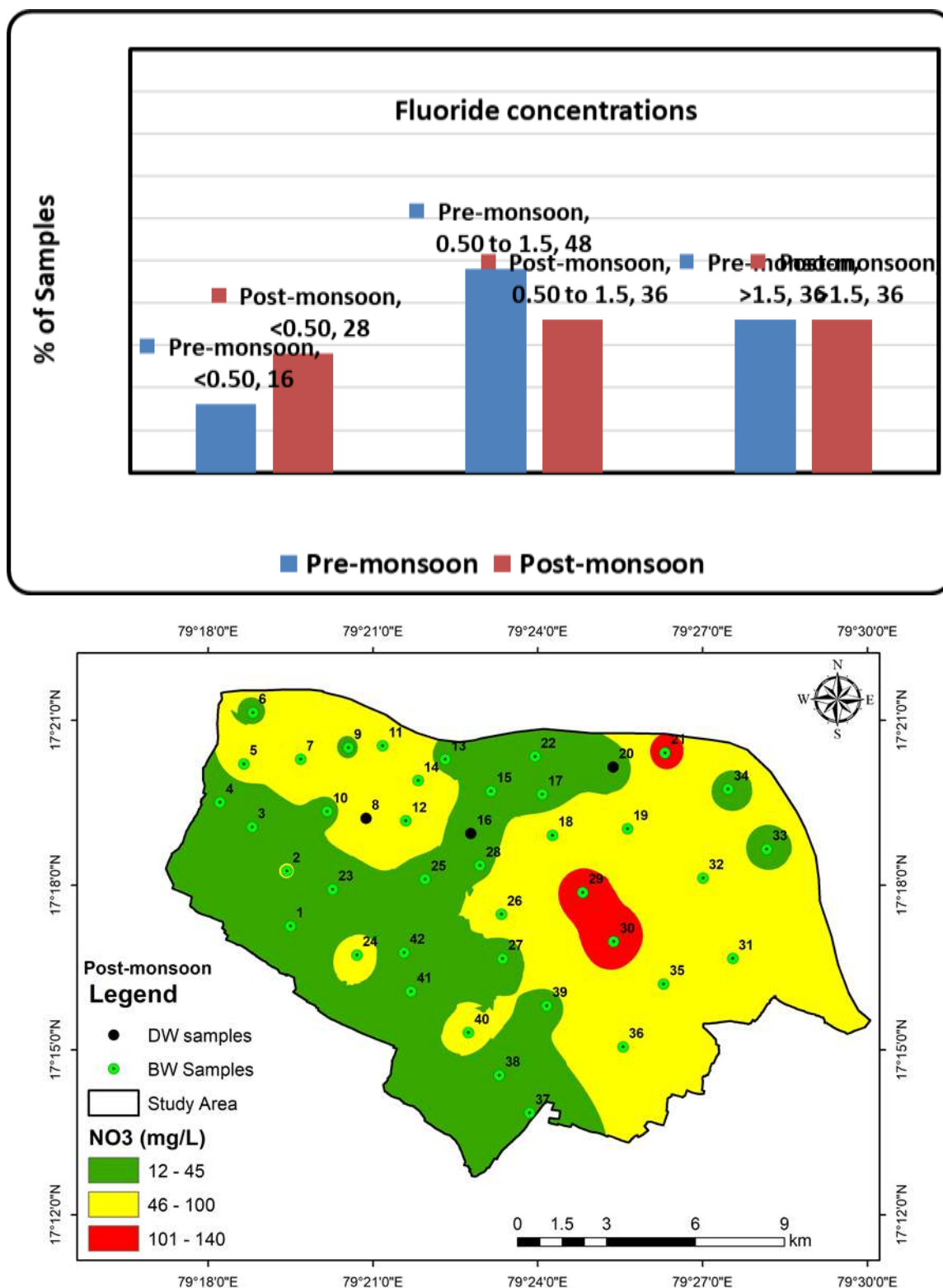


Fig. 6 Fluoride concentration levels in pre and post-monsoon seasons

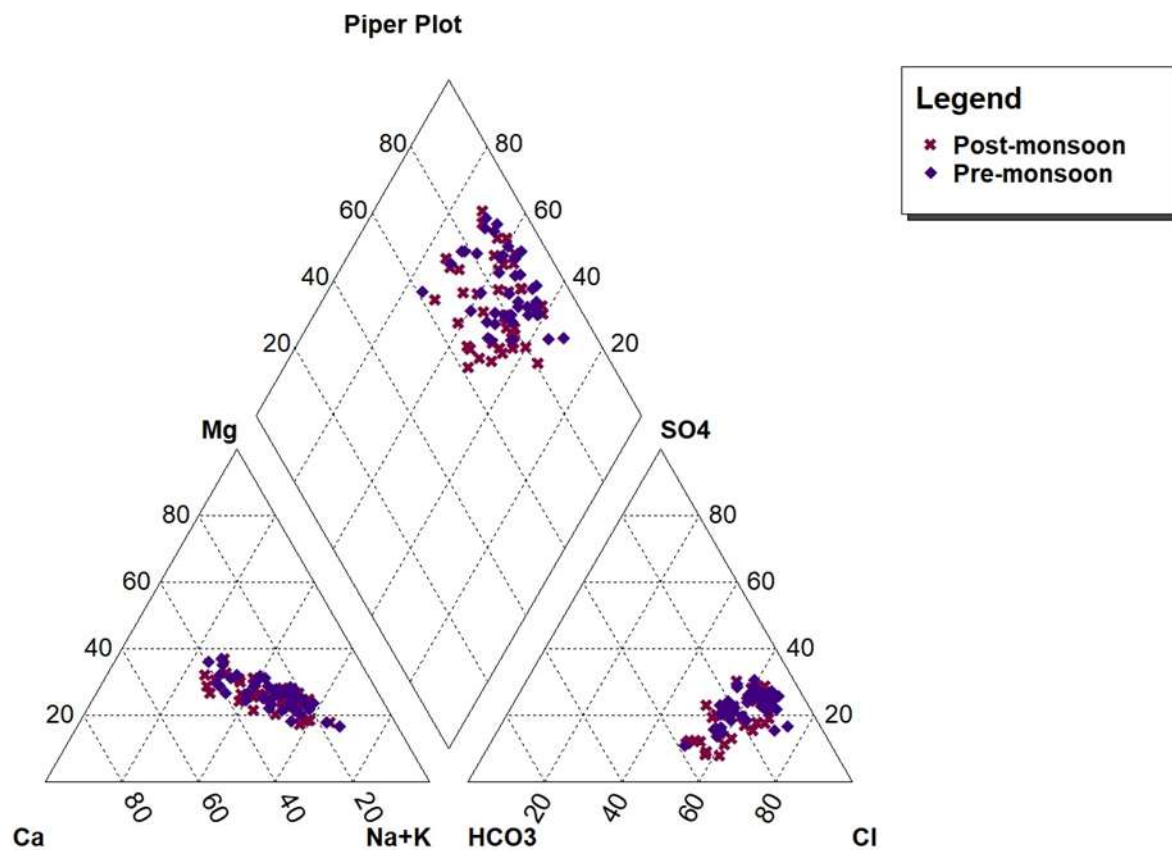


Fig. 7 Hydrochemical facies map of the study area

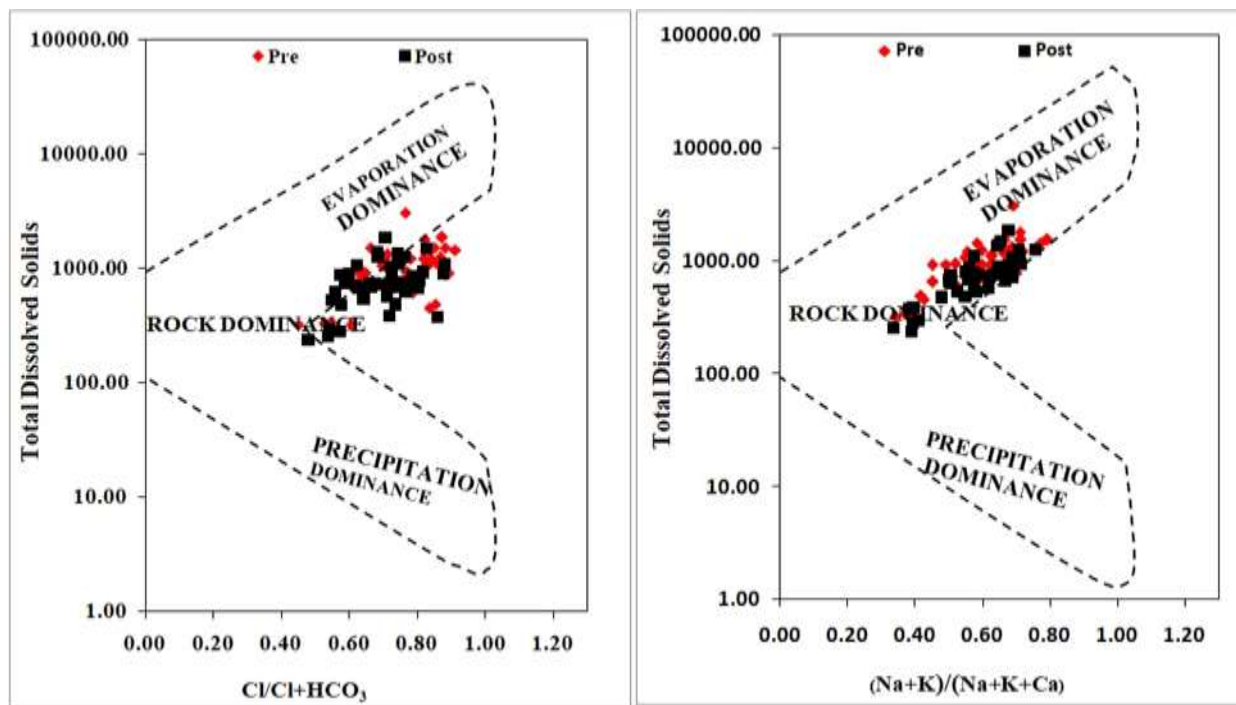


Fig. 8 Gibb's plot of the Study area

Table 1 Statistics of groundwater chemistry of the study area

Variables	Pre-monsoon season				Post-monsoon season				BIS (2012)
	Minimum	Maximum	Mean	% of samples exceeded the limits	Minimum	Maximum	Mean	% of samples exceeded the limits	
pH	7.80	9.40	8.26	12	7.56	9.22	8.28	21	6.5 - 8.5
EC ($\mu\text{S}/\text{cm}$)	495	4766	1623	-	400	2867	1197	-	-
F ⁻ (mg/l)	0.24	2.70	1.30	67	0.15	2.11	1.08	64	1.5
NO ₃ ⁻ (mg/l)	18	188	53	50	12	140	50	41	45
Gibb's I	0.45	0.91	0.75	-	0.48	0.88	0.69	-	-
Gibb's II	0.34	0.79	0.59	-	0.34	0.76	0.59	-	-