Occurrence of Uranium in Groundwater from Cuddalore District Tamil Nadu Aided by Geospatial and Statistical Techniques

P. Anandhan [a,*], S. Chidambaram [b], R. Manivannan [c], P. Paramaguru [a], C. Karthikeyan [c], K. Srinivasamoorthy [d], R. Prakash [d]

[a] Department of Earth sciences, Annamalai University, Annamalai Nagar-608002, Tamil Nadu, India.

[b] Research scientist, Water Research Center, Kuwait Institute for Scientific Research, Kuwait.

[c] Department of Geology, Alagappa Government Arts College, Karaikudi- 630003, Tamil Nadu, India.

[d] Department of Earth Sciences, Pondicherry University, Puducherry- 605014, India.

* Corresponding Author: vijayanand_pa@yahoo.co.in

Abstract: An attempt has been made to examine uranium distribution in groundwater from Cuddalore district, Tamil Nadu, India. Groundwater occurs under porous sedimentary, fractured, and weathered hard rock formations ranging in age from recent sediments to the oldest Archean formations. A total of 186 groundwater samples were collected during Pre-Monsoon (May) and Post-monsoon (January) and analyzed for major cations, anions, and uranium using standard procedures. Major anions and cations follow the order $Cl^- > H_4SiO_4 > HCO_3^- > NO_3^- > Na^+ > Ca^{2+} > Mg^{2+} > K^+ > SO_4^{2-} > F > PO_4^{3-}$ irrespective of seasons. Uranium in groundwater ranges from 0.1 micro gram per liter (μg/l) to 24.67 μg/l with average 1.82 μg/l. The spatial representation maps isolated areas of higher and lower uranium and statistical analysis inferred uranium sources to the groundwater environment.

1. Introduction

The groundwater chemistry is of greater importance in determining the suitability for utilities corresponding industrial, agricultural, and domestic utilities [1-4]. In coastal regions where groundwater being the primal source for clean water, like that of the study area, human interventions like over-drafting result in water quality degradation due to seawater intrusion and

related health issues. Uranium (U) is found in soil, water and humans in three isotopic forms (U-238, U-235, and U-234). U-238 and U-235 are the parent nuclides and the third isotope U-234 is the product of U-238 decay series. Uranium in the groundwater environment seems to be influenced by factors like lithology, geomorphology, and other environmental considerations of the study area. Uranium in groundwater is harmful to human exposure due to the chemical influence of aqueous hexavalent ions on the kidneys. Isolating uranium in water is of primal significance because of the hydrogeochemical significance and health risk assessment. Uranium in groundwater and its influence on human health has been attempted in India [5-10]. Sources of uranium to groundwater are mainly from mining regions, uraniferous conglomerates, and granitic intrusions [11-13]. For the present study, attempt has been made to focus on the spatial distribution of uranium occurrences in Cuddalore district of Tamilnadu, India, along with its geochemical significances aided by statistical analysis.

2. Study Area

The area demarcated for study is the Cuddalore district of Tamilnadu, India that lies between 15^0 05" and 12^0 35" N, 78^0 38" and 80^0 00" E with total coverage of about 3,678 Sq.km (Figure 1). The district is constrained by Villupuram district in north and northwest, Perambalur district due southwest, Ariyalur and Nagapattinam in the south and the Bay of Bengal along east. The study area is marked with pichavaram mangroves, Cuddalore port, and State Industries Promotion Corporation of Tamilnadu Ltd (SIPCOT) disseminated throughout the study area. The majority of the study area is influenced by agricultural practices that include paddy, cumbu, maize, varagu, blackgram, greengram, sugarcane, groundnut, gingelly and cotton plantations. The economically viable resources isolated in the study area were lignite, oil and natural gas. The annual average rainfall over the study area is 1160.12 nm, with limited showers during the southwest monsoon (between June to September) and significant rainfall during the northeast monsoon (October to January). The monthly mean temperature ranges from 40.34°C to 20.37°C. Rivers like Gadilam and Pennaiyar drains along with the northern parts of the study area, and Vellar and Coleroon drain and the study area's southern parts.

3. Geology ad Hydrogeology

The geology of the area gains importance in determining groundwater occurrences. This area is underlined by litho units ranging in age from Archean rocks to recent sediments. Tertiary and quaternary sedimentary rocks form the major litho units (Figure 2) covered by clay and clay sandstones, and exposure of limestone lenses, sandstone with mottled clay and lignite are found to cover significant portions of the study area. A small portion of Charnockite is exposed along with the northwestern parts of the study area. Groundwater in the study area is mainly confined to fractured and weathered Charnockites. In sedimentary formations, groundwater occurs in phreatic with confined to semi-confined conditions. The litho unit's groundwater level varies between 3.0 to 85.0 m Below Ground Level (BGL).
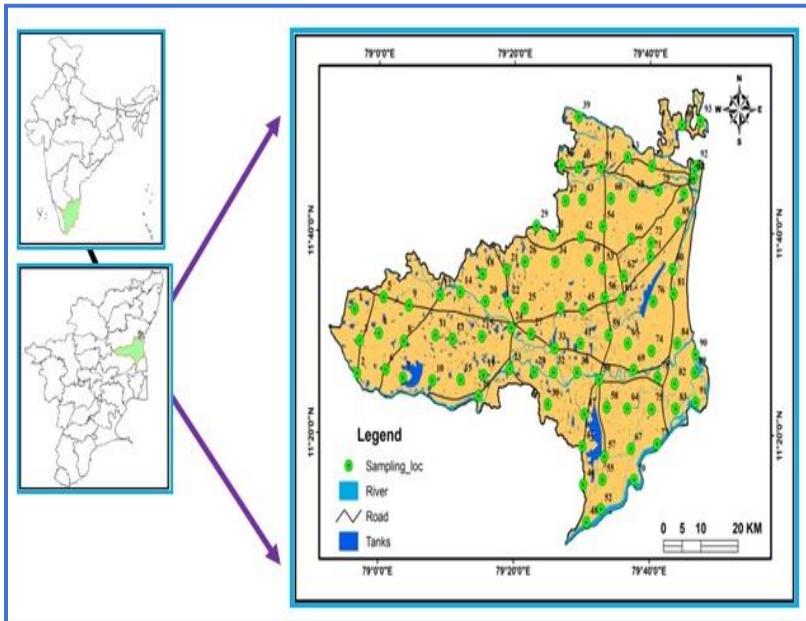
Figure 1. Location Map of the Study area
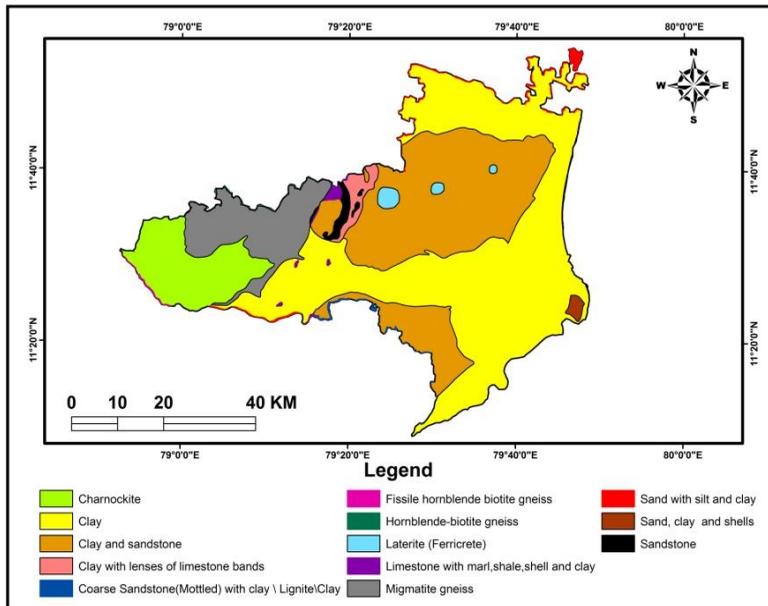
Figure 2. Geology Map of the study area.
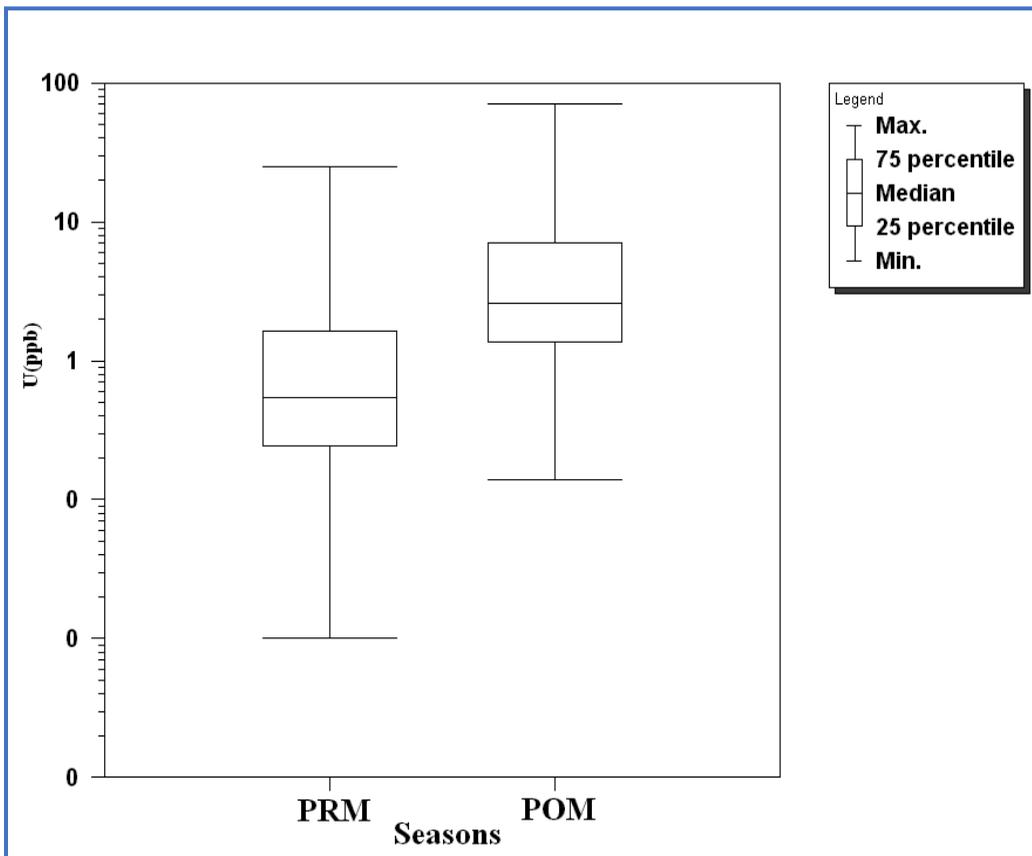
Figure 3. Box Plot for U in groundwater samples in different seasons

4. Result and Discussion

A total of 186 groundwater samples were collected and analyzed for two seasons' viz— pre-monsoon (PRM) and Post monsoon (POM). The maximum, minimum concentration, and seasonal variation of U in groundwater are represented as box plot (Figure 3).

The average U is found to increase with the monsoon, and a higher concentration was recorded during the POM season. Comparison of uranium with groundwater level indicates variation influenced by recharge and discharge areas. During rainfall, recharge to groundwater is significant, resulting in the high water table and increased uranium dissolution. Recharge of groundwater and its interaction with aquifer matrix releases uranium into the groundwater environment. After subsequent rainfall, during PRM uranium, decreases signifying a lesser role of recently recharged groundwater in the effective dissolution of uranium to the groundwater environment decreases.

4.1 Spatial variation of Uranium

The concentration of uranium was spatially represented during PRM and POM seasons. Uranium during PRM ranges between 0.01 µg/l to 24.67 µg/l. Higher uranium during PRM was confined to Southern parts of the study area, and lower were confined to eastern, southwestern, and northern parts of the study area (Figure 4a).

During POM season (Figure 4b), uranium ranges between 0.14 µg/l to 51.15 µg/l with higher concentration noted along with the southern parts of the study area, moderate and lower uranium were confined to southwestern and northeastern parts of the study area. Compared with PRM, more significant uranium was recorded during POM, suggesting influence from litho sources and anthropogenic influences. Changes in uranium are found to be influenced by pH and ORP. The permissible limit of U in drinking water by WHO, 2011 is 30.0 µg/l and the maximum acceptable limit as per Atomic Energy Regulatory Board (AERB, 2004) is 60.0 µg/l. Compared with the above standards, all the samples during PRM were within the permissible limit except 5 samples during POM that exceeded the WHO's permissible limit (2011) [14].
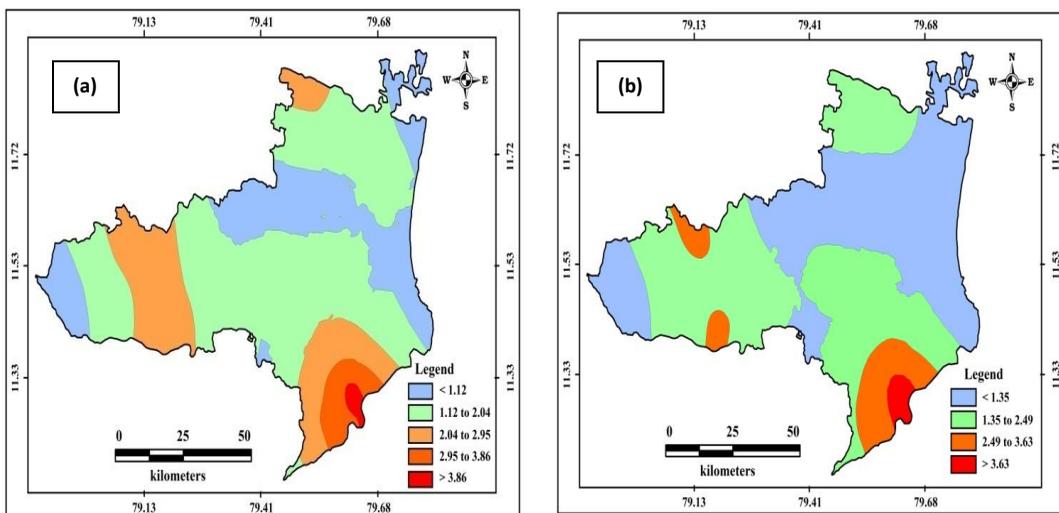
Figure 4. Spatial variation of U in groundwater samples a) PRM   b) POM seasons

4.2 Influence of pH on U

More excellent solubility of uranium is at pH 2.0 (U aqueous concentration ~0.01 M), which decreases with increasing pH to 4.0 (U aqueous concentration ~10^{-8} M), leveling out up to pH 8.0, and then uranium increases at pH 10.0 (U aqueous concentration ~10^{-6} M).As in the aqueous phase, most hexavalent U minerals contain U in the form of UO_2^{2+}. Variation of uranium at neutral pH is mainly due to strong complexation of uranyl ion with aqueous carbonate and the other complexate of uranyl ions are the phosphate and fluoride ions.

In general, the pH of water samples in the study area ranges between 5.5 to 7.9, representing acidic to alkaline. An increase in pH is mainly due to the mineral dissolution that increases uranium due to adsorption or reliable solution with the reacting media. The study area, an increase in pH is also found to increase uranium irrespective of seasons (Figure 5). Due to the influence of precipitation, uranium is varying with pH in the study area's groundwater. Higher uranium was observed at near-neutral pH, suggesting the adsorption of U is strongest above neutral pH values, and consequently, high pH values tend to affect the adsorption of U, which is again a function of pCO_2 of groundwater.
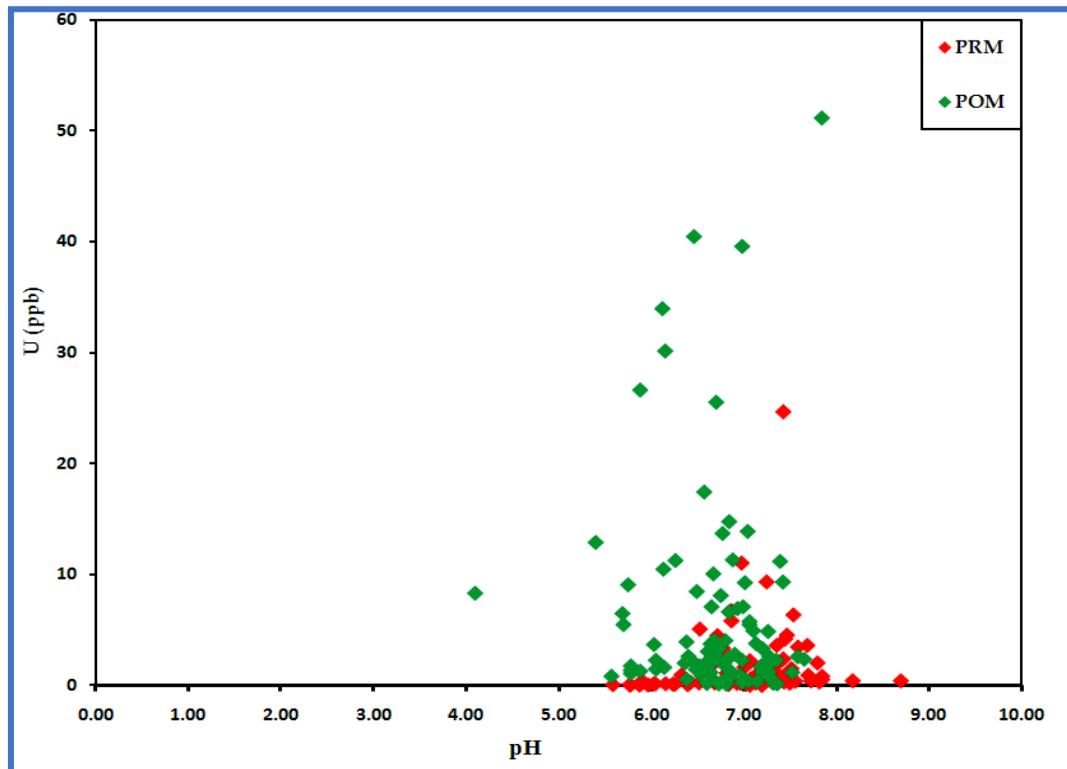
Figure 5. The plot of U vs. pH in groundwater samples for two seasons

4.3 Effect of U with EC

The association between U and EC is plotted in Figure 6. It is motivating to note that U is found to increase with greater EC irrespective of seasons, suggesting the influence of water residence time. The study area encompassed crystalline rock formations, with a more significant residence time of water due to cracks and fissures, and the presence of highly porous sedimentary formations increases the U concentration. The increase of EC may also be due to pH-tempted aquifer mineral weathering. U in groundwater diminished with flow direction due to U-retention by sorption processes.
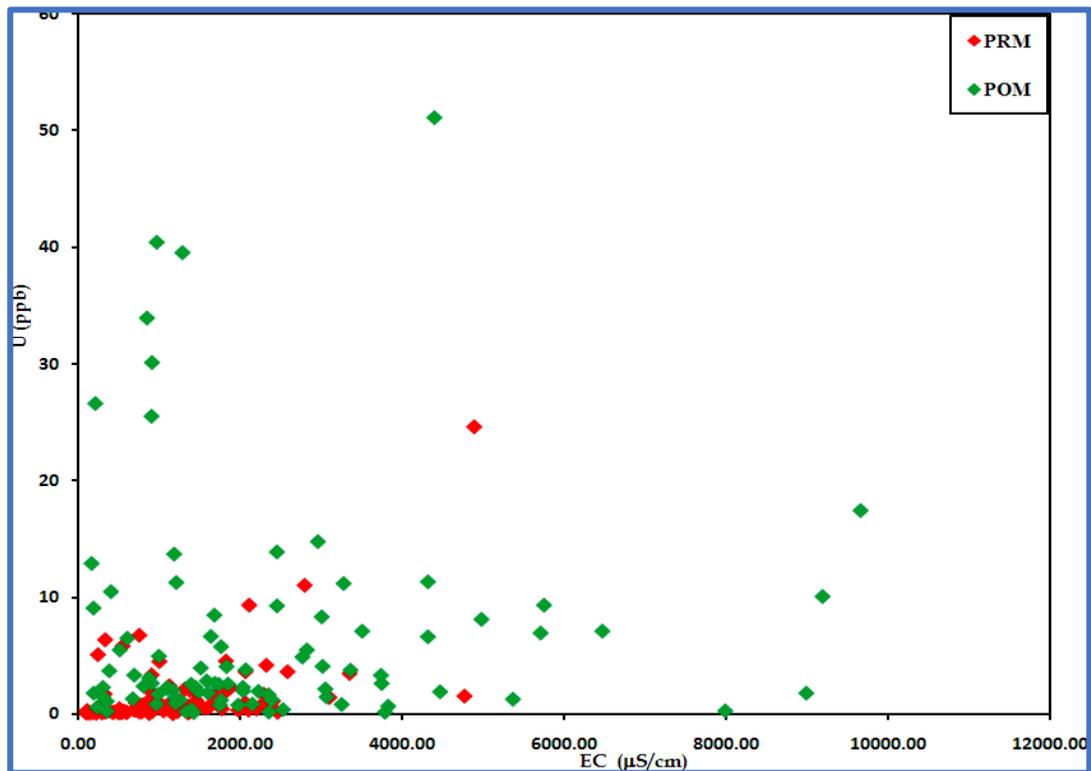
Figure 6. The plot of U vs. EC in groundwater samples for two different seasons

4.4 Effect of U with HCO_3^-

Higher bicarbonates (HCO_3) in groundwater is mainly due to the weathering of silicates aided by recent recharge waters [9]. The seasonal variations of HCO_3^- concerning U are represented in the plot (Figure 7). During PRM, HCO_3 is found to decrease along with uranium in most samples, and during POM, uranium increases with bicarbonate suggesting higher uranium and bicarbonate, signifying shallow groundwater conditions due to recent recharge.

4.5 Effect of pCO_2 with HCO_3^-

The pCO_2 plays a significant role in altering the solubility of carbonate rocks. Sources of pCO_2 are mainly by the dissolution of plagioclase minerals resulting in increased pH and mineral weathering. In the study area, the Log pCO_2 ranges from -0.35 to 1. (Figure 8), suggesting an increase of pCO_2 also increases bicarbonate.
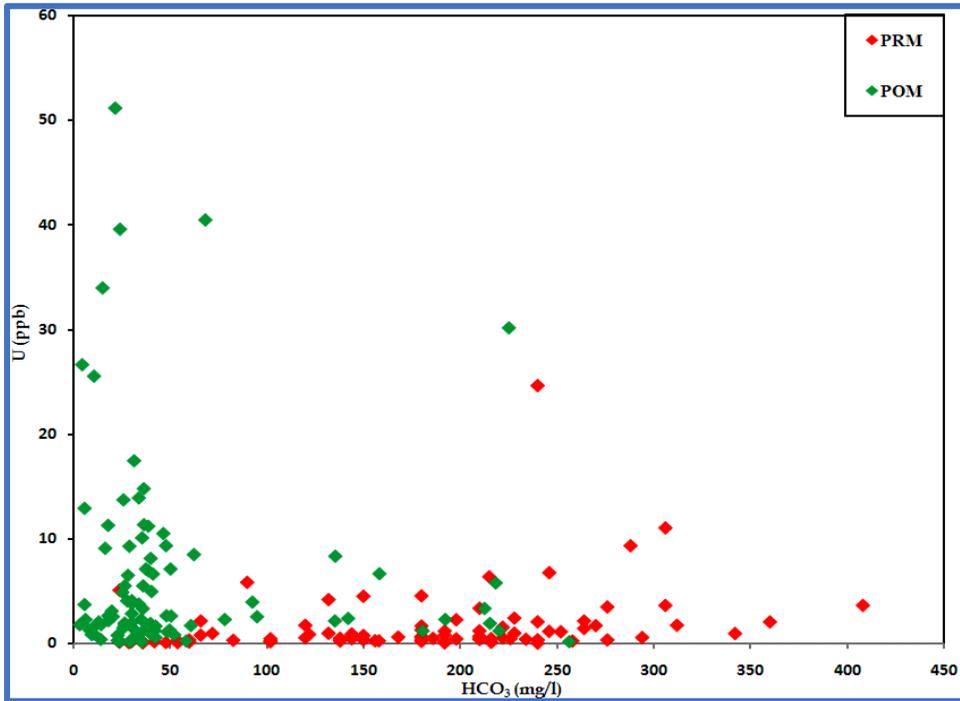
Figure 7. Plot of U vs. HCO3- in groundwater samples of all seasons
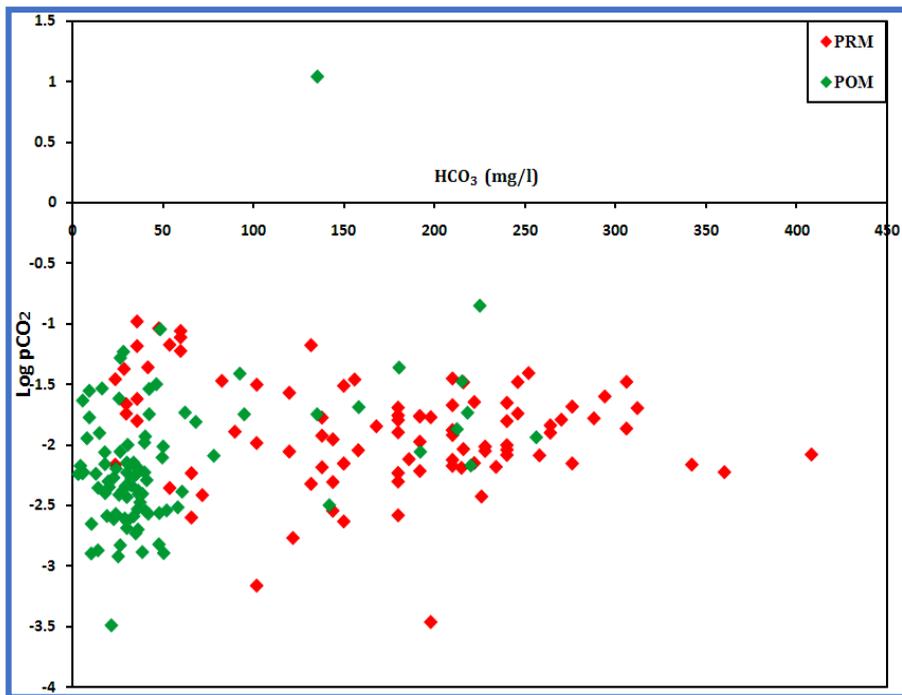
Figure 8. The plot of pCO2Vs HCO3 in groundwater samples irrespective of seasons

Chemical weathering of crystalline rocks plays a vital role in controlling the influence leaching phenomenon, dispersion and distribution of uranium minerals from the source rocks. Higher uranium with log pCO_2 values between -1 to -1.5 (Figure 9) suggests deeper groundwater circulation due to lower atmospheric interaction and or due to greater carbonate mineral saturation from aquifer matrix. The uranium source is mainly by weathering followed by uranium transportation aided by variation in pH and Bicarbonate concentrations.
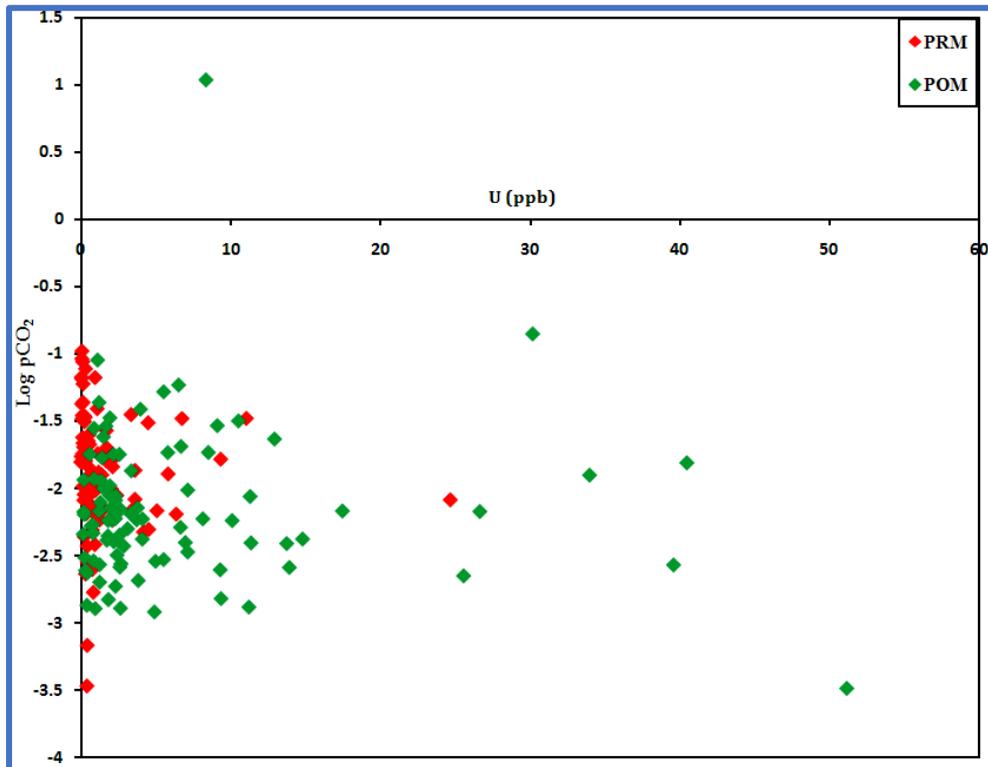
Figure 9. The plot of pCO2Vs U in groundwater samples irrespective of seasons

During PRM, good to excellent correlation is obtained between Ca^{2+}-Mg^{2+}, Cl^-, SO_4^{2-}; Mg^{2+} -Na^+, Cl^-; Na^+-Cl^-, U; K^+- PO_4^{3-}; HCO_3^--pH, EC; Cl^--SO_4^{2-} (Table 1), indicating the influence of weathering and leaching from the litho units of the study area. Cl shows a good correlation with Ca^{2+}, Na^+, and Mg^{2+} indicates leaching of secondary salts. A significant correlation of HCO_3^- with pH and EC indicates chemical weathering. A low positive correlation of SO_4^{2-}, NO_3^- with other ions might be due to the influence of dilution. A Positive, strong Uranium correlation with Mg and Na implies that uranium may be present in drinking water as a dissolved salt. pH shows a low positive correlation with U suggesting influence due to soil minerals' dissolution in the aqueous environment.

During POM, a good correlation exists between Cl^-- Mg^{2+}, Na^+; HCO_3^-- H_4SiO_4 (Table 2), a low correlation exists between Mg^{2+}, Na^+ with other ions. Cl^- shows a good correlation with Mg^{2+} and Na^+, suggesting sources from leaching of secondary salts from the study area's litho units and or due to industrial influences. Ions like Cl^- and HCO_3^- show a significant correlation with Ca^{2+}, Mg^{2+}, indicating the predominance of chemical weathering along with leaching of secondary salts. Nitrate shows good, moderate to a higher degree of correlation irrespective of seasons suggesting sources due to anthropogenic influences like the application of fertilizers.

Table 1. Correlation for groundwater samples collected during PRM

PRM	Ca^{2+}	Mg^{2+}	Na^+	K^+	F	Cl^-	HCO_3^-	SO_4^{2-}	PO_4^{3-}	NO_3^-	H_4SiO_4	U	pH	EC
Ca^{2+}	1.0													
Mg^{2+}	.56	1.00												
Na^+	.36	.58	1.00											
K^+	.19	.25	.30	1.00										
F	-.21	.06	.21	-.08	1.00									
Cl^-	.70	.73	.79	.37	-.09	1.00								
HCO_3^-	.20	.41	.40	-.01	.41	.08	1.000							
SO_4^{2-}	.16	.44	.48	.09	.13	.40	.32	1.00						
PO_4^{3-}	-.04	-.14	.05	.54	-.09	.00	-.02	-.09	1.00					
NO_3^-	.03	.16	.17	.14	.17	.01	.30	.02	.06	1.00				
H_4SiO_4	.16	-.06	-.04	-.07	-.01	.02	.00	.21	-.04	.06	1.00			
U	.30	.51	.55	.10	.03	.47	.27	.320	-.03	.09	-.06	1.00		
pH	.05	.24	.33	.10	.23	.07	.61	.262	.01	.15	-.00	.13	1.00	
EC	.26	.44	.55	.10	.08	.34	.57	.45	.00	.17	-.04	.48	.41	1.00

Table 2. Correlation for groundwater samples collected during POM

POM	Ca^{2+}	Mg^{2+}	Na^+	K^+	F	Cl^-	HCO_3^-	SO_4^{2-}	PO_4^{3-}	NO_3^-	H_4SiO_4	U	pH	EC
Ca^{2+}	1.00													
Mg^{2+}	.34	1.00												
Na^+	.24	.34	1.00											
K^+	.18	.13	.24	1.00										
F	-.05	.08	.25	.04	1.00									
Cl^-	.42	.52	.94	.24	.22	1.00								
HCO_3^-	.22	.28	.28	-.00	.08	.30	1.00							
SO_4^{2-}	-.02	.12	.18	.08	-.09	.19	.03	1.00						

PO_4^{3-}	-.19	.00	.18	-.07	-.08	.13	.11	.00	1.00					
NO_3^-	.12	.15	.18	.38	.07	.15	-.10	-.03	-.05	1.00				
H_4SiO_4	.08	.04	.00	-.05	-.01	-.01	.501	-.02	.15	-.170	1.00			
U	-.07	-.02	.10	.12	.34	.08	-.05	.05	.04	.05	-.05	1.00		
pH	-.137	.00	.13	-.03	.29	.06	.20	-.09	.23	.01	.11	.01	1.00	
EC	.12	.20	.47	.26	.18	.44	-.03	.14	-.04	.43	-.12	.04	.25	1.00

4.7 Factor Analysis

Factor analysis (**FA**) is a widely used statistical technique in hydrochemical research to explain geochemistry of groundwater along with demarcation of hydrochemical facies. FA is also applied to investigate sources of groundwater contamination.

The usual interpretation of groundwater's chemical quality using ionic ratio plots for significant ions does not define the similarities between ions or samples. Factor analysis is a powerful tool to detect similarities among the variables or samples. Factor analysis interprets the samples' structure by extracting the eigenvalues and eigenvectors from the correlation or covariance matrix. The factors are constructed to reduce the data's overall complexity by taking advantage of inherent inter-dependencies resulting in a small number of factors that give the same information as those of larger data sets. The interpretation is based on rotated factors, rotated loadings, and rotated eigenvalues. Factor analysis does not require prior knowledge of the number of sources influencing the samples, nor does it require knowledge of the source composition.

Factor analysis has been attempted for **PRM** and **POM** data sets. The sorted FA results along with loading of variables, eigen values and data set variances were represented by individual factors for two different seasons. The factor loadings were sorted according to the criteria [15], i.e., substantial, modest, and weak, corresponding to absolute loading values of >0.75, 0.75–0.50, and 0.50–0.30, respectively.

4.7.1 Pre Monsoon

FA rendered four significant factors that explain about 64.8% of Total Data variability (Table.3). The ions in Factor I show a total variance of about 29.48%, indicating the influence of high EC and the dominance of Ca^{2+}, Mg^{2+}, Na^+, Cl^-, SO_4^{2-}, and U, indicating leaching of secondary salts may be due to industrial influences (Table 3). This factor clearly isolates Na^+ and Cl^- are mainly from saline sources. Factor II was represented with a variance of 16.5% influenced by ions like F, HCO_3^-, pH, and EC. HCO_3^- and Fs loading is due to the high HCO_3^- bearing water having alkaline nature, which favours F ions' higher mobility in the groundwater.

Factor III recorded with a variance of about 11%, representing K^+ and PO_4^{3-}, indicates anthropogenic sources due to residential water softeners' application, sources from septic tanks,

or fertilizers application during agricultural practices weathering of potash feldspar process. Factor IV enriched with the H_4SiO_4 representing 7.7%TDV dissolution of silicate minerals. Factor IV shows the negative loadings of Na^+, K^+, F, PO_4^{3-}, and pH.

4.7.2 Post Monsoon

FA extracted four significant factors during the post-monsoon season, representing 64.76% of the total variance (Table 4). Factor 1 represented a total variance of 20.3% by ions like Na^+, K^+, Cl^-, NO_3^-, and EC, indicating sources from secondary salt leaching. Factor II extracted with a total variance of about 14.6% (Table 4), suggesting enrichment of ions like Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^- and H_4SiO_4 due to intensive weathering of silicate minerals. Factor III represented a data variance of about 10.2 % influenced by ions like PO_4^{3-} and pH (Table 4), suggesting impact due to fertilizer applications. With a total variance of 10.0%, Factor IV is represented by Cl^- and SO_4^{2-} signifying sources due to fertilizers' application. Factor V extracted with a total variance of 9.8% represented by U and F ions suggesting similar chemical control.

Table 3. Factor analysis for PRM samples (Varimax rotated)

PRM	1	2	3	4
Ca^{2+}	.69	-.22	.05	.32
Mg^{2+}	.81	.12	-.00	.02
Na^+	.79	.30	.16	-.07
K^+	.26	-.04	.84	-.01
F	-.09	.64	-.13	-.05
Cl^-	.91	-.17	.16	.08
HCO_3^-	.30	.80	-.04	.02
SO_4^{2-}	.54	.32	-.10	.24
PO_4^{3-}	-.11	.02	.86	-.06
NO_3^-	.01	.45	.27	.23
H_4SiO_4	-.02	.02	-.06	.92
U	.68	.12	-.05	-.19
pH	.18	.70	.07	-.01
EC	.59	.50	.00	-.10
TDV%	29.40	16.50	11.00	7.70

Table 4. Factor analysis for POM samples (Varimax rotated)

POM	1	2	3	4
Ca^{2+}	.26	.52	-.54	.02
Mg^{2+}	.26	.52	-.24	.26
Na^+	.56	.42	.14	.50

K^+	.58	-.01	-.21	.01
F^-	.17	.12	.08	-.17
Cl^-	.53	.52	-.01	.53
HCO_3^-	-.08	.82	.14	-.02
SO_4^{2-}	.01	-.06	.02	.73
PO_4^{3-}	-.04	.09	.70	.24
NO_3^-	.69	-.14	-.14	-.19
H_4SiO_4	-.24	.62	.23	-.19
U	-.03	-.13	-.04	.14
pH	.23	.17	.66	-.32
EC	.81	-.00	.15	.08
TDV%	20.00	146.00	10.20	10.00

4.8 Factor Score

The Factor scores are projections of data onto corresponding eigenvectors that provide information about the factors' placement. Factor scores were attempted for the present study by adopting the regression technique. The positive zones demarcate the dominance of that particular factor in influencing the study area's hydrogeochemical regime.

The first factor for **PRM** and **POM** seasons was spatially plotted to gain information about the first factor's active zone (Figure 10 and 11). Factor 1 during **PRM** is confined to the central parts of the study area, dominant with clay and sandstone litho units suggesting sources due to leaching of secondary salts and industrial influences. Factor 2 is found to dominate along with the eastern and western parts of the study area, with dominant litho units encompassing clay sandstone, charnockite, and agricultural land use. Factor 3 and 4 represent the eastern and central parts of the study area influenced by agricultural activities and sources due to seawater intrusion confined to the sedimentary environment.

During POM, Factor 1 is represented in the eastern parts of the study area consisting of sedimentary (clay and sandstone) formations, representing influence due to secondary leaching and saline water intrusion. Factor II is dominant along the northern and western regions of the study area, nearly made up of agricultural land use with dominant litho units encompassing gneiss, clay, and sandstone formations. Factor III was noted in the central parts of the study area with the dominance of clay and limestone litho units and influenced by agricultural practices. Factor VI is confined to eastern parts of the study area composed of clay and sandstone formations, suggesting influence due to anthropogenic activities.
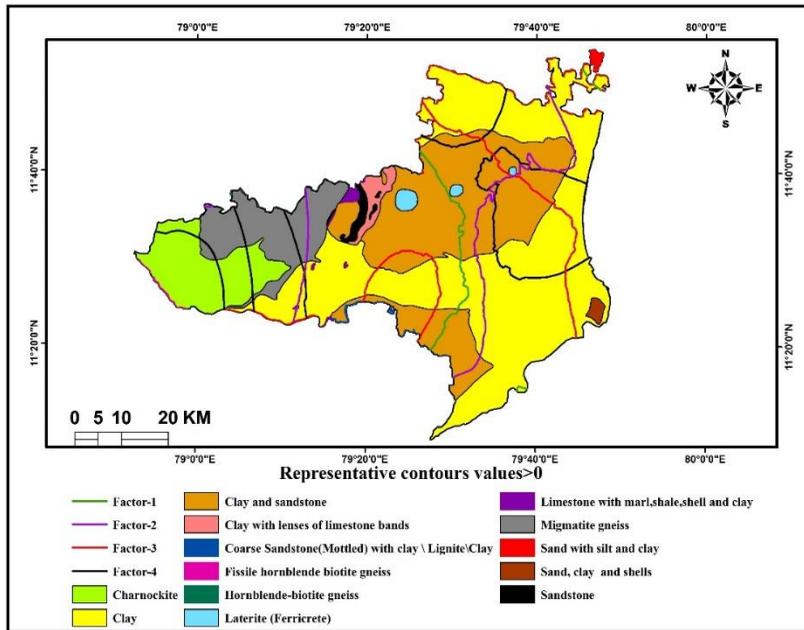
Figure 10. Spatial distribution of factor score correlated with Lithology during PRM in groundwater
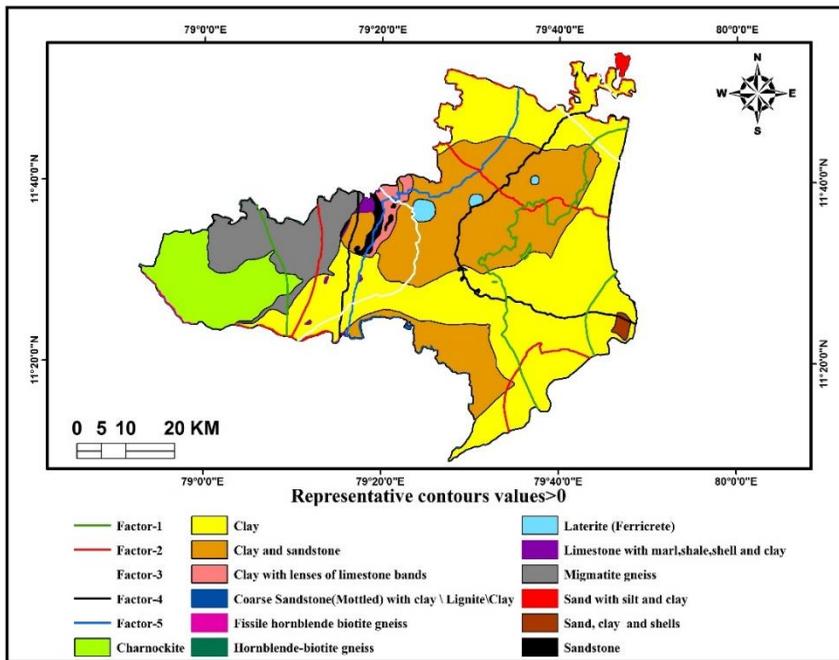
Figure 11. Spatial distribution of factor score correlated with Lithology during POM in groundwater

In general, the dominance of secondary leaching, saline water intrusion, and anthropogenic related activities seem to influence the study area's groundwater chemistry. Positive representation of factor 1 is prevalent along with the northwestern, central, and southern parts of the study area [16].

Due to the complex hydrological scenario, it is impossible to extract all the factors responsible for altering the study area's hydrochemistry. Hence, five factors were extracted for a total of two different seasons (PRM and POM). The first factor signifies the dominance of secondary salts dissolution. Second, third, fourth, and fifth factors record anthropogenic activities' influence (Figure 10 and 11).

5. Conclusion

The concentration of uranium shows that it increases with the monsoon. Higher U noted in POM seasons suggests sources from litho units along with anthropogenic influences. Higher uranium is mainly due to weathering followed by uranium transport aided by variation in pH and bicarbonate variations irrespective of seasons. Higher uranium during PRM was confined to Southern parts of the study area, and lower were confined to eastern, southwestern, and northern parts of the study area. Statistical analysis suggests the dominance of secondary leaching, anthropogenic influences like agricultural and industrial related activities, and seawater intrusion to control the study area's geochemistry irrespective of seasons.

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