



High background radiation places and spatial distribution of uranium in groundwater of monazite placer deposit in Kanniyakumari district, Tamil Nadu, India

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Abstract

The present investigation deals with gamma radiation survey and analysis of uranium in groundwater samples collected from the Kanniyakumari district. It reveals that gamma dose rate varies from 58 to 3880 nSv/h (mean 276 nSv/h) and uranium concentration varies from ≤ 0.2 to 10 $\mu\text{g/L}$ (median 0.4 $\mu\text{g/L}$) in the pre-monsoon and ≤ 0.2 to 14.9 $\mu\text{g/L}$ (median 0.2 $\mu\text{g/L}$) in the post-monsoon seasons. The present study shows that Manavalakurichi and Indranagar junctions have high background radiation due to the monazite placer deposits. The alkaline and reductive nature of groundwater minimizes the leaching of uranium from these monazite placer deposits.

Keywords Dose assessment · Monazite deposits · Saturation Index · Uranium in groundwater · Water Quality Index

Introduction

Water is one of the essential ingredients for the survival of life. Uranium is a naturally occurring radioactive element present in all the environment's compartments, including the hydrosphere, with wide variation. Uranium is present in minerals like uraninite, pitchblende, carnallite, complex oxides of silicates, phosphates, vanadates, lignite, and monazite sands [1]. These radioactive minerals emit radiation continuously due to primordial radionuclides of ^{40}K , ^{238}U , and ^{232}Th . These radioactive minerals containing places are known as High background radiation places and reported in many countries like Nigeria, Brazil, Germany, and India [2]. In India, Kerala and Kanniyakumari district's coastal regions in Tamilnadu have been reported for high background radiation due to the scattered deposits of monazite sand [3–7].

The leaching of natural deposits and anthropogenic activity is responsible for higher uranium concentration in groundwater [8–10]. The acidic or alkaline medium and oxidizing condition of groundwater favors the dissolution of uranium in groundwater. Nitrate pollution and water table decline incite the uranium mobilization in groundwater [11, 12]. The reported epidemiological and laboratory studies show that uranium primarily causes health problems via poisoning the kidney, bone, liver, reproductive system, lung, and nervous system. The studies showed that the long-term ingestion of elevated uranium levels in drinking water causes chronic health effects such as nephrotoxicity, bone toxicity, reproductive toxicity, hepatotoxicity, neurotoxicity, and pulmonary toxicity [13]. Considering the health concern of uranium ingestion, the World Health Organization (WHO) and the Atomic Energy Regulatory Board (AERB) of India have set the guideline values as 30 $\mu\text{g/L}$ and 60 $\mu\text{g/L}$, respectively, for uranium in drinking water [14–16]. Uranium concentration has been reported in groundwater with 3410 $\mu\text{g/L}$ in Finland and 1442 $\mu\text{g/L}$ in Karnataka, India [17, 18]. This high uranium in Finland and Karnataka is due to the geogenic sources, mainly granitic rocks. The presence of uranium in the groundwater has been controlled by hydrolysis reactions (precipitation, complexation, etc.) with anions (carbonate, bicarbonate, sulphate, chloride, etc.) and cations (sodium, potassium, calcium, magnesium, etc.). The

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oxidized form of uranium, U(VI), travels long distances than the reduced form U(IV) in the groundwater [19].

In the Kanniyakumari district, people depend on groundwater for their drinking and domestic purposes. The southwest coast of the Kanniyakumari district is facing groundwater contamination problems due to natural factors and anthropogenic sources such as removal of sand, coastal encroachment, seawater intrusion, sea-level rise. Many authors have reported the groundwater quality of the Kanniyakumari district [20–23], but to our knowledge, the studies on the concentration of uranium in groundwater are not reported. This district has high background radiation due to the occurrence of monazite minerals. In this concern, the gamma radiation survey was performed to estimate the dose exposure of outdoor gamma radiation. Uranium concentration in groundwater was measured to evaluate groundwater quality under WHO and AERB standards. The present study aims to assess the uranium sources and local hydrogeochemical conditions on implication with the groundwater resources.

Materials and methods

Study area

Kanniyakumari district is on the southern border of India and situated in between 77° 9' 32.71" E to 77° 35' 18.67" E longitude and 8° 4' 37.01" N to 8° 16' 44.83" N latitude. It covers a small area of 1672 km² but has a high population density (1119/km²), second in Tamil Nadu. It has about 71 km of coastal line, the Gulf of Mannar on the east, the Indian Ocean on the south, and the Arabian Sea on the west. Kanniyakumari has a border with districts, Tirunelveli in the northeast, Thiruvananthapuram (Kerala) in North West, and the Western Ghats in the North. The study area is classified into two regions, Edai Nadu (Vilavancode and Kalkulam taluks) and Nanjil Nadu (Thovalai and Agastheeswaram taluks) [20, 24].

Red, Red lateritic, Brown, and coastal soils are the main soil types in the Kanniyakumari district. Lateritic soils are present in Vilavancode and Kalkulam taluks and mixed types of alluvial and red soils in Agastheeswaram and Thovalai taluks. The Southwest coastal line contains soils with garnet, illuminite, and monazite minerals [24, 5]. The study area has various geological units such as charnockite and garnet–biotite gneiss, sandstone associated with clay materials, and granites. Sand, silt, and clay units are distributed predominantly compared with other geological units. The district's major geological formation is peninsular gneiss, and gabbro formation is the least predominant one (Fig. 1). It receives rainfall from two seasons; southwest monsoon (June–September) and northeast monsoon

(October–December), with an annual rainfall of 80–241 mm. The main drainage is the Pazhayar river flowing from the Western Ghats to the southern direction [24]. The people depend on open-well for their freshwater needs. Water scarcity arises when the southwest monsoon (June–September) and northeast monsoon (October–December) fail.

Gamma radiation survey

Thirty-nine stations (Table 1) were selected to assess the background radiation and measure the concentration of uranium in groundwater (Fig. 2). The gamma radiation survey was conducted in the selected stations using the hand-held radiation survey meter (Polimaster-1405). It operates with the Geiger-Miller counter principle and gives a precise measurement of gamma radiation dose rate in the range from 0.01 nSv/h to 100 mSv/h [25]. The outdoor gamma dose rates were measured by the survey meter at one meter height from the ground level. From the height of 1 m measurements, the activity concentrations of gamma radiation in the air are emitted by the radioactive isotopes present in the different rock types in the sampling villages. The measurements were carried out at one-meter height above the ground level, and at least six meters away from any building or walls is to avoid effects of ground and buildings on outdoor measurements [26]. The 10 min average values of the gamma dose rate were taken in each location. The coordinates of the sampling stations were recorded using the Garmin GPS device [27, 28].

Sample collection and analysis

Sixty-eight groundwater samples were collected in 1 L HDPE (High-Density Polyethylene) bottles during the pre-monsoon (September–October-2016) and post-monsoon (January-2018) seasons. The bottles were pre-rinsed with concentrated nitric acid and completely washed with distilled water. The bottles were pre-rinsed with the samples before collecting the water samples. After collecting the samples, a few drops of concentrated HNO₃ was added to preserve the samples, and it is stored in an ice-box (4 °C) [29]. The physical parameters pH, total dissolved solids, electrical conductivity, and oxidation–reduction potential of the groundwater were measured in the field using the hand-held water quality analysis kit (BWASA-5 Biotech portable digital water & soil analysis kit). The primary anions, bicarbonate, carbonate, chloride, and the primary cations calcium, magnesium were analyzed by the titrimetric method using high-grade chemicals. The sodium and potassium ions were analyzed using the flame photometer (EI, model No. 381E), and the remaining anions of chloride, nitrate, phosphate were analyzed by the Spectrophotometer

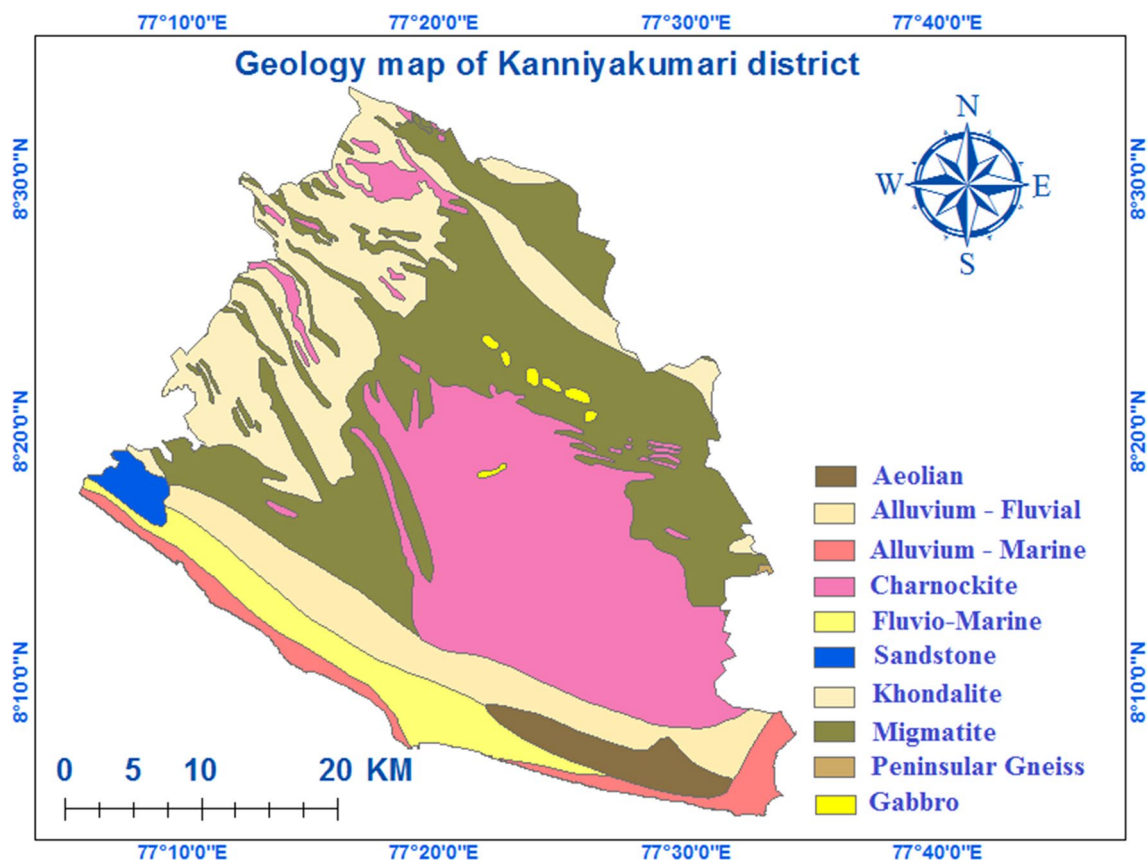


Fig.1 Geology map of the study area

(Elico SL-177). The fluoride concentration was analyzed using an ion-selective electrode (HI 4110) [30–33].

Uranium measurements

The concentration of uranium in the groundwater samples was measured using the LED-Fluorimeter LF-2a, which operates on the principle of Beer-Lambert law. Samples were treated with phosphate buffer (pH=7) to form a fluorescent uranyl-phosphate complex measured at 405 nm [30, 34]. For minimizing the error in the measurements, the standard addition method was employed [29]. The standard addition method's detection limit is 0.2 ppb, and the instrument has a detection range of 0.1–1000 µg/L. The accuracy of the measurements is $\pm 5\%$ [30–32].

Water quality index (WQI)

The equation $WQI = \sum_{i=1}^n SI_i$ is used to calculate the water quality index using the analyzed parameters, where SI_i is the sub-index of i th parameter calculated from $SI_i = W_i \times Q_i$. Here, Q_i ($C_i/S_i \times 100$) is the quality rating of i th parameter calculated from the concentration of the parameters (C_i) and

their guideline values (S_i), and W_i is the relative weight calculated from assigned weight w_i (Table 2) [35, 36].

Interpretation of data

For data interpretation, spatial distribution maps were plotted using GIS software version 10.3. Hydrogeochemistry of the groundwater was studied using the piper plot (plotted using Aqua chem. software ver. 4) and Gibbs plot (plotted in the MS Excel spreadsheet). The saturation indices of the minerals were evaluated using the PHREEQC interactive 2.8 software.

Results and discussion

High background radiation places

The measured gamma dose rates vary from 58 to 3880 nSv/h with a mean of 276 nSv/h (Fig. 3). Previous studies show a similar outdoor background gamma radiation level 120–3780 nSv/h in the coastal area of Kanniyakumari district [16]. The annual effective dose equivalents were calculated by the equation [37],

Table 1 Station name and location of the stations

S. no	Station name	Latitude	Longitude
S1	Kappukadu	8.2935	77.1961
S2	Melpuram	8.3431	77.2164
S3	Karod	8.4036	77.2268
S4	Kotrakonam	8.4238	77.2101
S5	Kannamamoodu	8.3762	77.1694
S6	Kaliakkavilai	8.3311	77.1752
S7	Alangi	8.1951	77.2350
S8	Indranagar Junction	8.2034	77.2205
S9	Kuchiparavilvai	8.2189	77.2120
S10	Ayunivilai	8.2394	77.1789
S11	Alangodu	8.2840	77.1561
S12	Krathoor	8.2839	77.1289
S13	Manavalakurichi	8.1520	77.306
S14	Thuckalay	8.2396	77.3340
S15	Saralvilai	8.2693	77.3184
S16	Ponmanai	8.3542	77.3283
S17	Pechiparai Dam	8.4493	77.3083
S18	Thirparappu	8.3947	77.2653
S19	Thiruvattar	8.3431	77.2769
S20	Karugankuzhi	8.2151	77.2711
S21	Thumbanthottam	8.2792	77.2661
S22	Nullivilai	8.2034	77.3469
S23	Kallukootam	8.1868	77.2890
S24	Surulode	8.3399	77.3753
S25	Muttam	8.1247	77.3180
S26	Chungunkadai	8.2020	77.3815
S27	Vembanur	8.1696	77.3709
S28	Ganapathipuram	8.1313	77.3608
S29	Kottar	8.1800	77.4376
S30	Puthenthurai	8.1023	77.4194
S31	Manakudi	8.0932	77.4896
S32	Kanyakumari	8.0841	77.5500
S33	Maharajapuram	8.1259	77.5430
S34	Valukkamparai	8.1524	77.4832
S35	Thovalai	8.2258	77.4992
S36	Aralvoimozhi	8.2450	77.5191
S37	Erichakulam	8.2276	77.4289
S38	Chiramadam	8.2884	77.4316
S39	Thadikarakonam	8.3234	77.4197

$$\text{AEDE (Outdoor) (mSv/y)} = \text{Absorbed dose (nSv/h)} \\ \times 0.7 \text{ (conversion coefficient)} \times 8760 \text{ h} \\ \times 0.2 \text{ (fraction of spent time in outdoor)} \times 10^{-6}.$$

The calculated AEDE values vary from 0.07 to 4.76 mSv/y, with a mean of 0.34 mSv/y (Table 3). This average annual outdoor effective dose of Kanniyakumari

district (0.34 mSv/y) is within the recommended national (India) and worldwide limit of 1 mSv/y [38, 39]. This district's previous studies show that the AEDE values range from 0.05 to 1.25 mSv/y with a mean of 0.46 mSv/y [17]. The study region's value is anticipated as placer deposits having 1–2% of monazite mineral containing thorium and uranium and their daughter products. However, the coastal stations of Kanniyakumari district, Manavalakurichi (S-13, 2 mSv/y), and Indranagar junction (S-8, 4.76 mSv/y) have higher annual effective dose rate due to the heterogeneous distribution of monazite in the region. The high radiation level is due to the coastal soils containing heavy minerals like ilmenite, rutile, garnet, zircon, monazite, magnetite, sillimanite, etc. The rich deposits of monazite-bearing black sand slicks consist of orthophosphate of thorium, uranium, and rare earth elements. The weathering of mountains carried through estuaries into the ocean is also one reason for this high background radiation [40].

Water quality parameters

The statistical results and spatial distribution maps of the analyzed water quality parameters are presented in Table 3 and supplementary Fig. 1. From the hydrogeochemical data of both seasons, pH values are within the BIS permissible limit of 6.5–8.5 [41]. Most water samples are alkaline, with an average value of 7.1 (pre-monsoon) and 7.7 (post-monsoon). This alkaline nature may be due to the dissolution of carbonate rocks such as calcite or dolomite [35]. About 36% (pre-monsoon) and 28% (post-monsoon) of the samples have Total Dissolved Solids (TDS) higher than the BIS permissible limit of 500 ppm [41].

The literature shows that Manakudi (S-31) sampling location is in the high vulnerability zone (southeast) of seawater intrusion. A higher TDS concentration of 1560 mg/L is recorded in the sample collected from Manakudi (S-31), demonstrates that the seawater intrusion may be responsible for the higher TDS in the sample [21–23, 42]. Kanyakumari district receives rainfall from both the southwest and southeast monsoon, and therefore there is no drastic change in groundwater level in pre-monsoon (2.66–20.06 m bgl), post-monsoon (1.19–14.57 mg/L), and also many surface water sources (Lake, Pool, etc.). This type of higher rainfall and availability of surface water is directly related to the groundwater sources. In the present study, lower TDS values are noted in the collected samples due to the availability of surface water sources, which reduces groundwater extraction leads to poor seawater intrusion in this district [21–24, 42]. The collected water samples' electrical conductivity varies from 25 to 3110 $\mu\text{S/cm}$ in pre-monsoon and from 31 to 3496 $\mu\text{S/cm}$ in post-monsoon. The oxidation–reduction potential (ORP) values range from –57 to 26 mV in pre-monsoon and –78 to 8 mV in post-monsoon. These negative

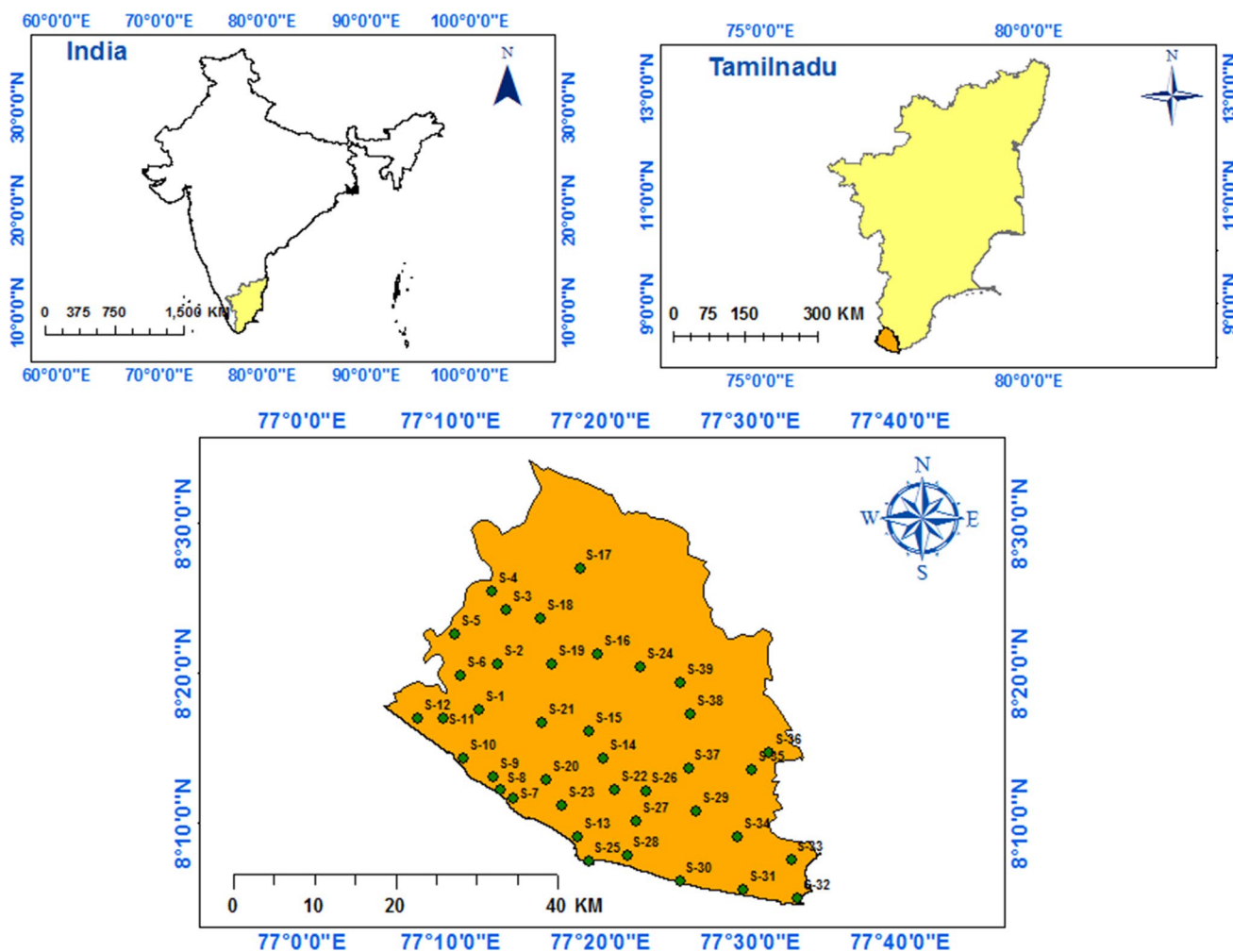


Fig.2 Map of the study area

Table 2 Classification of ground water based on water quality index

Water quality index		Number of samples		Percentage of samples	
Range	Quality of water	Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon
< 50	Excellent	24	24	62	62
50–100	Good	10	13	26	34
100–200	Poor	4	1	10	2
200–300	Very poor	1	1	2	2
> 300	Unsuitable	–	–	–	–
	Total	39	39	100	100

values of ORP indicate that the groundwater in the study area is in reducing condition [43].

The average anion concentration follows the order of $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{PO}_4^{3-} > \text{F}^- > \text{CO}_3^{2-}$ in the pre-monsoon and $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{CO}_3^{2-} > \text{F}^- > \text{PO}_4^{3-}$ in the post-monsoon. In both seasons, HCO_3^- ion predominates with the variation of 165 to 2149 mg/L, with an average of 511 mg/l in pre-monsoon and 80 to

1316 mg/L in post-monsoon, with an average of 352 mg/L in post-monsoon. Fluoride ion concentration in the analyzed samples varies from 0.1 to 0.8 mg/L in the pre-monsoon, and 0.1–1.2 mg/L in post-monsoon, and all the samples are within the BIS permissible limit of 1 ppm [41] except one sample from post-monsoon (Kottar (S-29), 1.2 mg/L). Nitrate concentration is also within the permissible limit of 45 ppm in all the samples during both

Fig.3 Spatial distribution of background gamma radiation in pre-monsoon (PRM) (nSv/h)

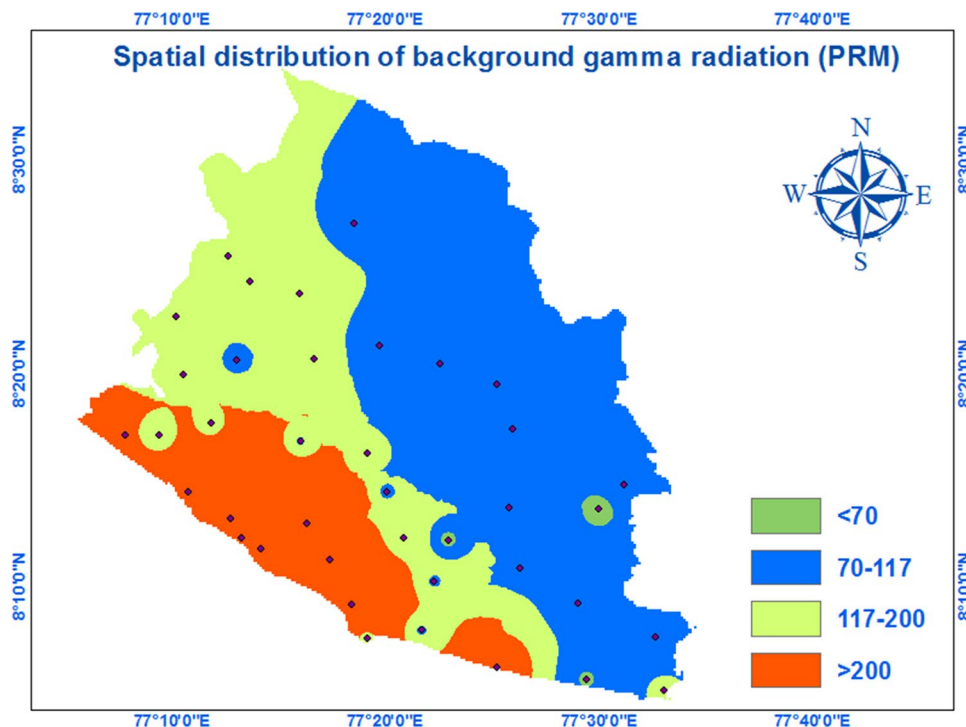


Table 3 Minimum, maximum and average values of water quality parameters and gamma radiation

Parameters	Pre-monsoon			Post-monsoon			BIS 2012
	Min	Max	Average	Min	Max	Average	
pH	6.5±0.1	8.1±0.1	7.1±0.1	6.8±0.1	8.4±0.1	7.7±0.1	6.5–8.5
TDS (mg/L)	13±0.1	1560±0.1	455±0.1	16±0.1	1840±0.1	402±0.1	500
EC (µS/cm)	25±0.1	3110±0.1	897±0.1	31±0.1	3496±0.1	775±0.1	–
ORP (mg/L)	–57±1	26±1	–6±1	–79±1	8±1	–37±1	–
F [–] (mg/L)	0.1±0.1	0.8±0.1	0.4±0.1	0.1±0.1	1.2±0.1	0.4±0.1	1.0
Cl [–] (mg/L)	21	1053	200	8	1263	189	250
NO ₃ [–] (mg/L)	0.5	9.3	1.1	1	6	1	45
SO ₄ ^{2–} (mg/L)	2	1373	81	1	1112	79	200
PO ₄ [–] (mg/L)	0.1	2.7	0.6	0.1	1.1	0.3	–
CO ₃ ^{2–} (mg/L)	0	0	0	0	32	1	–
HCO ₃ [–] (mg/L)	165	2149	624	80	1316	428	–
Alkalinity (mg/L)	136	1762	511	66	1078	352	–
Na ⁺ (mg/L)	5	532	80	2	558	74	–
K ⁺ (mg/L)	0.1	2.3	1	0.1	2.2	1.2	–
Ca ²⁺ (mg/L)	0	189	52	0	187	47	75
Mg ²⁺ (mg/L)	10	76	31	5	75	26	30
Gamma radiation level (nSv/h)	58±5	3880±5	276±5	–	–	–	1
U (µg/L)	0.1±0.05	10.5±0.05	2±0.05	0.1±0.05	14.9±0.05	1.6±0.05	60 (AERB)

seasons. On chloride concentration, 31% (pre-monsoon) and 21% (post-monsoon) of the samples exceed the permissible limit of 250 ppm, with an average of 200 mg/L in pre-monsoon and 180 mg/L in post-monsoon. Sulphate concentration varies from 2 to 1373 mg/L with an average

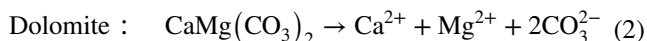
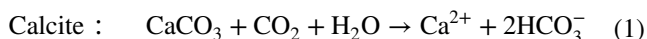
of 81 mg/L in the pre-monsoon and 1–1111 mg/L with 79 mg/L in the post-monsoon seasons. Vembanur (S-27) is the only station that exceeds the permissible sulphate limit (200 mg/L).

The average cations in the analyzed samples in both seasons follow the order $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$. Na^+ is predominant in both seasons with a variation of 5–532 mg/L with an average of 80 mg/L in pre-monsoon and 2–558 mg/L with 74 mg/L in post-monsoon. About 31% and 26% of the samples exceed the permissible limit of calcium (70 mg/L) during pre-monsoon and post-monsoon, with an average value of 52 mg/L and 47 mg/L, respectively. On magnesium concentration, 35% and 33% of the samples exceed the permissible limit of 30 mg/L with an average of 31 mg/L and 26 mg/L during pre- and post-monsoon. The potassium concentration varied from 0.1 to 2.3 mg/L, with an average of 1 mg/L in pre-monsoon and 0.1–2.2 mg/L with 1.2 mg/L in post-monsoon.

Correlation matrix of the pre-monsoon (Table 4) water quality parameters exhibit strong positive ($r > 0.8$) correlation between the parameters, TDS with EC, Cl^- , Na^+ and EC with Cl^- , Na^+ and Cl^- with Na^+ and Ca^{2+} with Mg^{2+} . There are moderate correlation ($0.5 < r < 0.8$) between TDS with PO_4^{3-} , HCO_3^- , K^+ , Ca^{2+} , Mg^{2+} and EC with PO_4^{3-} , HCO_3^- , K^+ , Ca^{2+} , Mg^{2+} and Cl^- with PO_4^{3-} , K^+ , Ca^{2+} , Mg^{2+} and PO_4^{3-} with Na^+ and HCO_3^- with Na^+ , Ca^{2+} , Mg^{2+} and Na^+ with K^+ , Ca^{2+} , Mg^{2+} . There is a strong negative correlation between the pH and ORP with a value of -0.852. Correlation matrix of the post-monsoon (Table 5) samples show a strong positive correlation ($r > 0.8$) between TDS with Cl^- , Na^+ , Mg^{2+} and EC with Cl^- , Na^+ , Mg^{2+} and Na^+ with Cl^- and HCO_3^- with Ca^{2+} and Ca^{2+} with Mg^{2+} . The moderate correlation ($0.5 < r < 0.8$) between the parameters, TDS with HCO_3^- , Ca^{2+} and EC with SO_4^{2-} , HCO_3^- , Ca^{2+} and SO_4^{2-} with Cl^- , HCO_3^- , Na^+ , Ca^{2+} , Mg^{2+} and HCO_3^- with F^- , Na^+ , Mg^{2+} and Na^+ with Ca^{2+} , Mg^{2+} . A strong negative

correlation exists between the pH and ORP with a value of -0.997.

HCO_3^- is the predominant anion, and Ca^{2+} and Mg^{2+} are the predominant cations after the Na^+ ion. A moderate correlation between these parameters implies that the calcite, dolomite and aragonite weathering is possible in groundwater aquifer by the following Eqs. (1) and (2) [44, 45]:



The saturation index (Table 6) values of the calcite and dolomite in pre-monsoon (-0.2 and 0) and post-monsoon (0.2 and 0.7) are nearly zero confirms that the minerals are in equilibrium with the ions in groundwater.

Hydrogeochemical facies

The piper diagram is plotted for the samples to identify the hydrogeochemical facies of the groundwater. The piper plots drawn for the pre-monsoon water samples (Fig. 4a, b), demonstrate that the calcium-bicarbonate ($\text{Ca}^{2+} + \text{HCO}_3^-$), sodium-chloride ($\text{Na}^+ + \text{Cl}^-$) and mixed calcium-magnesium-chloride (mixed $\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Cl}^-$) water types are present with percentage of 72, 15 and 13, respectively [46]. The post-monsoon samples have the water types mentioned earlier with the percentage of 68, 28, 2, and the remaining 2% of the samples fall under the mixed $\text{Ca}^{2+} + \text{Na}^+ + \text{HCO}_3^-$ water types. About 72% of the pre-monsoon samples and 62% of the post-monsoon samples have weak acids ($\text{CO}_3^{2-} + \text{HCO}_3^-$) exceed the strong acids ($\text{SO}_4^{2-} + \text{Cl}^-$) and the remaining 28% of the pre-monsoon

Table 4 Correlation matrix for water samples of pre-monsoon (*The bold characters are the correlation values higher than 0.5)

	pH	TDS	EC	ORP	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	U
pH	1														
TDS	0.170	1													
EC	0.192	0.997	1												
ORP	-0.852	-0.143	-0.167	1											
F ⁻	-0.216	0.099	0.099	0.014	1										
Cl ⁻	0.132	0.938	0.936	-0.096	-0.005	1									
NO ₃ ⁻	-0.049	0.127	0.149	0.112	-0.029	0.102	1								
SO ₄ ²⁻	0.016	0.478	0.477	0.013	-0.037	0.467	0.028	1							
PO ₄ ⁻	0.435	0.553	0.551	-0.399	-0.083	0.586	-0.149	-0.041	1						
HCO ₃ ⁻	0.443	0.679	0.689	-0.480	0.290	0.471	0.090	0.313	0.442	1					
Na ⁺	0.205	0.930	0.930	-0.197	-0.002	0.984	0.094	0.490	0.608	0.523	1				
K ⁺	-0.002	0.595	0.603	0.012	-0.219	0.515	0.306	0.302	0.266	0.408	0.542	1			
Ca ²⁺	0.244	0.745	0.753	-0.263	0.154	0.626	0.144	0.463	0.378	0.666	0.629	0.293	1		
Mg ²⁺	0.298	0.781	0.792	-0.375	0.106	0.669	0.150	0.434	0.436	0.798	0.681	0.403	0.854	1	
U	0.327	0.766	0.785	-0.319	0.058	0.650	0.102	0.478	0.423	0.713	0.680	0.573	0.800	0.729	1

Table 5 Correlation matrix for water samples of post-monsoon (*The bold characters are the correlation values higher than 0.5)

	pH	TDS	EC	ORP	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ⁻	CO ₃ ²⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	U
pH	1															
TDS	0.174	1														
EC	0.175	1.000	1													
ORP	-0.997	-0.184	-0.185	1												
F ⁻	-0.091	0.204	0.205	0.095	1											
Cl ⁻	0.160	0.946	0.944	-0.162	0.114	1										
NO ₃ ⁻	-0.224	0.230	0.228	0.210	0.163	0.102	1									
SO ₄ ²⁻	0.231	0.581	0.578	-0.232	0.423	0.544	0.080	1								
PO ₄ ⁻	0.036	0.179	0.176	-0.026	-0.007	0.166	0.119	-0.065	1							
CO ₃ ²⁻	0.421	0.129	0.136	-0.457	-0.167	0.017	-0.062	0.016	-0.068	1						
HCO ₃ ⁻	0.025	0.690	0.691	-0.027	0.543	0.528	0.192	0.531	0.120	0.100	1					
Na ⁺	0.193	0.955	0.953	-0.196	0.153	0.967	0.162	0.533	0.263	0.054	0.630	1				
K ⁺	0.028	0.486	0.486	-0.024	0.111	0.402	0.338	0.228	0.108	-0.220	0.435	0.458	1			
Ca ²⁺	0.145	0.776	0.778	-0.161	0.469	0.627	0.277	0.676	0.182	0.206	0.822	0.720	0.443	1		
Mg ²⁺	0.105	0.810	0.814	-0.109	0.478	0.736	0.141	0.573	-0.012	0.188	0.790	0.755	0.341	0.814	1	
U	0.267	0.796	0.793	-0.256	0.333	0.786	0.124	0.746	0.270	0.035	0.653	0.811	0.292	0.716	0.669	1

samples and 38% of the post-monsoon samples have strong acids (SO₄²⁻ + Cl⁻) exceed the weak acids (CO₃²⁻ + HCO₃⁻). About 85% of the pre-monsoon samples and 92% of the post-monsoon samples have alkaline earth (Ca²⁺ + Mg²⁺) exceed the alkalis (Na⁺ + K⁺), and the remaining 15% of the pre-monsoon samples and 8% of the samples have the alkalis (Na⁺ + K⁺) exceed the alkaline earth (Ca²⁺ + Mg²⁺).

Gibbs plot represents the key process that controls the surface water chemistry. It is also used to explain the hydro-geochemical process that occurs in the groundwater. There are three natural processes: precipitation dominance, evaporation dominance, and rock–water interaction dominance [47]. Gibbs plot (Fig. 5a, b) of the present study shows that thirty-four samples in pre-monsoon and thirty-five samples in post-monsoon have the rock–water dominance and the remaining five samples in pre-monsoon and four post-monsoon samples have precipitation dominance [48]. In the study area, rock-water dominance is a key procedure that controls the chemistry of groundwater.

Water quality index (WQI)

The water quality index is useful to simplify the interpretation of many variables influencing water quality. In the collected samples, the water quality index was computed showed a higher percentage of uranium, fluoride, and nitrate, which will create health risks among the people consuming the water. Calcium, sulphate, magnesium, chloride, and TDS were also taken for the WQI calculation due to drinking water’s organoleptic characteristics. In the present study, the following classifications are made from the calculated water quality index values; excellent (< 50), good (50–100), poor (100–200), very poor (200–300), and unsuitable for drinking (> 300) [49]. In the samples, 88% (pre-monsoon) and 96% (post-monsoon) have WQI values within 100 belong to the excellent and good water category. 10% and 2% of the samples have poor water quality with the WQI values between 100 and 200 during the pre- and post-monsoons. The remaining 2% have very poor water quality, with the WQI values between 200 and 300 (Table 7). Totally 12% and 4% of the samples have poor and very poor water qualities in the pre and post-monsoon seasons. This implies that drinking this poor quality water will cause organoleptic effects due to elevated levels of chloride, calcium, magnesium, and TDS [49].

Uranium in groundwater

Uranium concentration in the analyzed samples varies from ≤ 0.2 to 10.5 µg/L (median 0.4 µg/L) in the pre-monsoon and ≤ 0.2–14.9 µg/L (median 0.2 µg/L) in the post-monsoon (Fig. 6a, b). The entire samples collected from the Kanniyakumari district have the uranium concentration

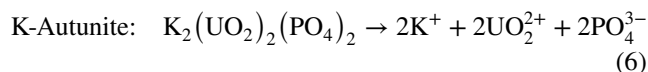
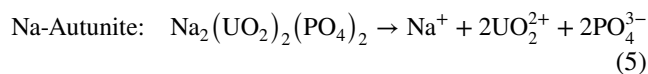
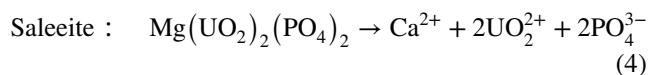
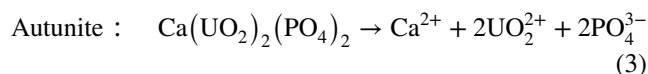
Table 6 Saturation index values of minerals in water samples of Kanniyakumari district

Minerals	Pre-monsoon (SI values)			Post-monsoon (SI values)		
	Min	Max	Average	Min	Max	Average
Anhydrite	-5.2	-0.9	-2.9	-5.5	-0.9	-3.0
Aragonite	-2.6	1.4	-0.3	-1.8	1.5	0.1
Calcite	-2.4	1.5	-0.2	-1.7	1.7	0.2
Dolomite	-3.5	3.1	0.0	-2.0	3.4	0.7
Fluorite	-4.3	-1.4	-2.6	-4.8	-1.0	-2.8
Gypsum	-5.0	-0.7	-2.7	-5.3	-0.7	-2.8
Halite	-8.5	-4.9	-6.9	-9.3	-4.8	-7.1
Uraninite	-30.1	-27.7	-28.8	-30.3	-26.7	-28.7

within the permissible limit of AERB (60 µg/L) and guideline value of WHO (30 µg/L) [8, 9]. The maximum uranium concentration is 10.5 µg/L (Puthenthurai, S-30) in pre-monsoon and 14.9 µg/L (Vembanur, S-27) post-monsoon. In the present study, higher radiation places, Manavalakurichi (U, 0.4 and 0.2 ppb) and Indranagar junction (U, 3.0 and <0.2 ppb) have low uranium concentration during both the seasons. But, high uranium concentration places, Puthenthurai (U, 10.5 ppb, pre-monsoon) and Vembanur (U, 14.9 ppb, post-monsoon) have low background radiation i.e., 334 nSv/h (Puthenthurai) and 405 nSv/h (Vembanur) when compared with the Manavalakurichi (1680 nSv/h) and Indranagar junction (3880 nSv/h). Puthenthurai and Vembanur have the background radiation ten times lesser than the Indranagar Junction (3880 nSv/h), but these two stations have a uranium concentration ten times higher than the Indranagar Junction. Gamma radiation levels were measured in the present study reflects the surface mineralogy, while the uranium level in groundwater is due to rock-water interaction and underlying geology and geochemistry. Therefore, it is difficult to correlate the ambient gamma radiation levels and uranium in groundwater when the surface and deeper rock composition is dissimilar and heterogeneous. When the surface and deeper layers have similar geology, then a positive correlation may be anticipated.

In present study the correlation of the parameters in pre-monsoon, shows uranium has strong positive correlation with Ca^{2+} ($r > 0.8$), moderate correlation ($0.5 < r < 0.8$) with Na^+ , K^+ and weak correlation ($r < 0.5$) with PO_4^{3-} . It illustrates that the secondary uranium minerals such as autunite and saleeite

in the monazite placer deposits of the Kanniyakumari district may get weathered under these reducing conditions (Eqs. 3–6). Sandstone and sedimentary rocks of the study area consist of these secondary uranium minerals [50, 51].



From the correlation matrix of post-monsoon, uranium has a strong positive correlation with Na^+ ($r > 0.8$), moderate correlation with K^+ ($0.5 < r < 0.8$) and weak correlation with PO_4^{3-} ($r < 0.5$). The correlation between uranium with other water quality parameters has varied from pre-monsoon to post-monsoon. The groundwater recharge can occur by the rainfall with the leached minerals is responsible for this variation in the collected pre- and post-monsoon water samples. The acidic and oxidizing conditions of groundwater favor the dissolution of uranium in groundwater. The groundwater's alkaline and reductive nature favor diminishing uranium concentration by the hydrolysis reactions with anions and cations present in the groundwater. The oxidized form of uranium U(VI) has high mobility than the reduced form U(IV) in groundwater [19]. In the present study, the oxidation–reduction

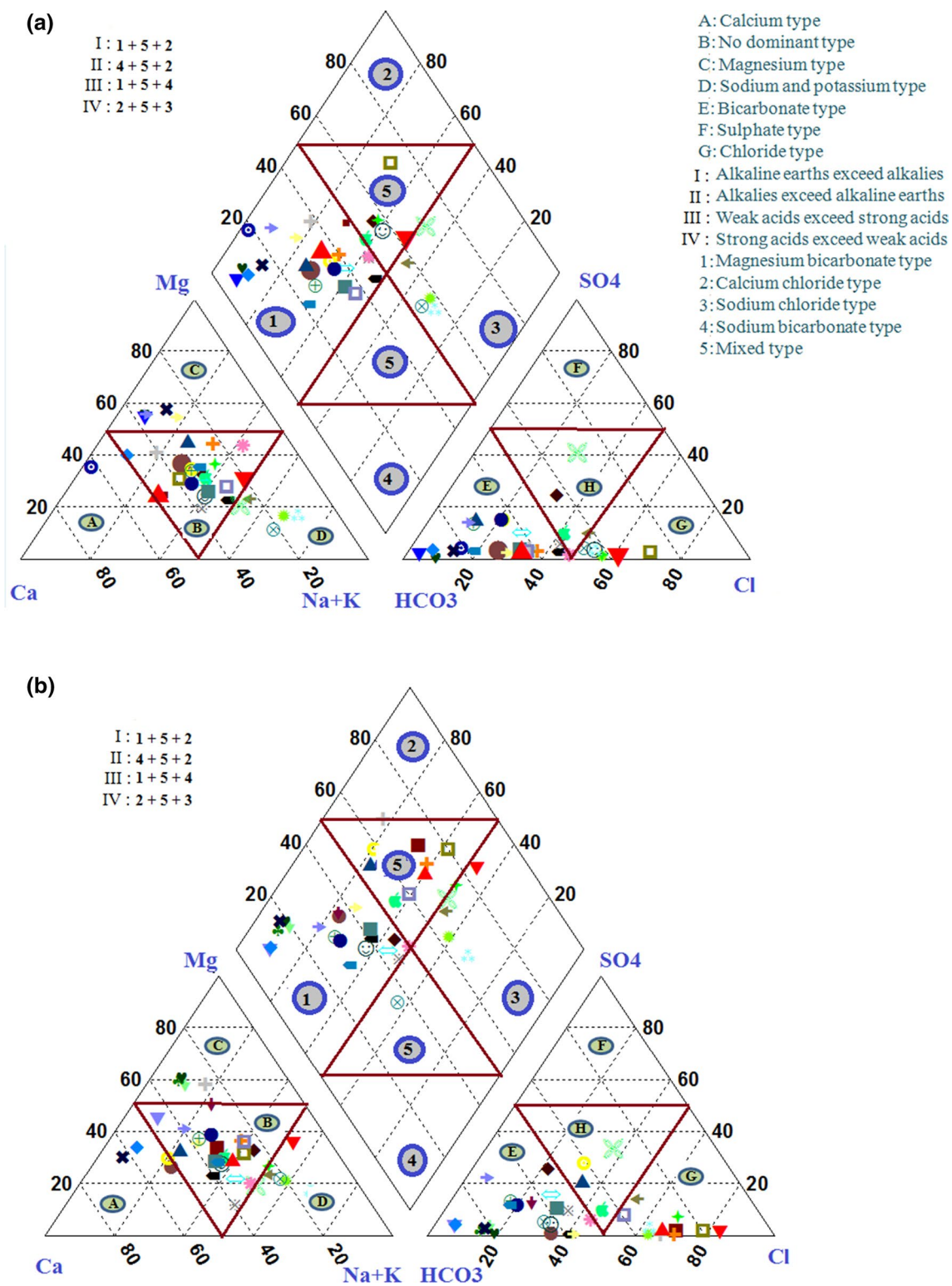


Fig.4 Piper plot for water samples from kanniyakumari district a pre-monsoon b post-monsoon

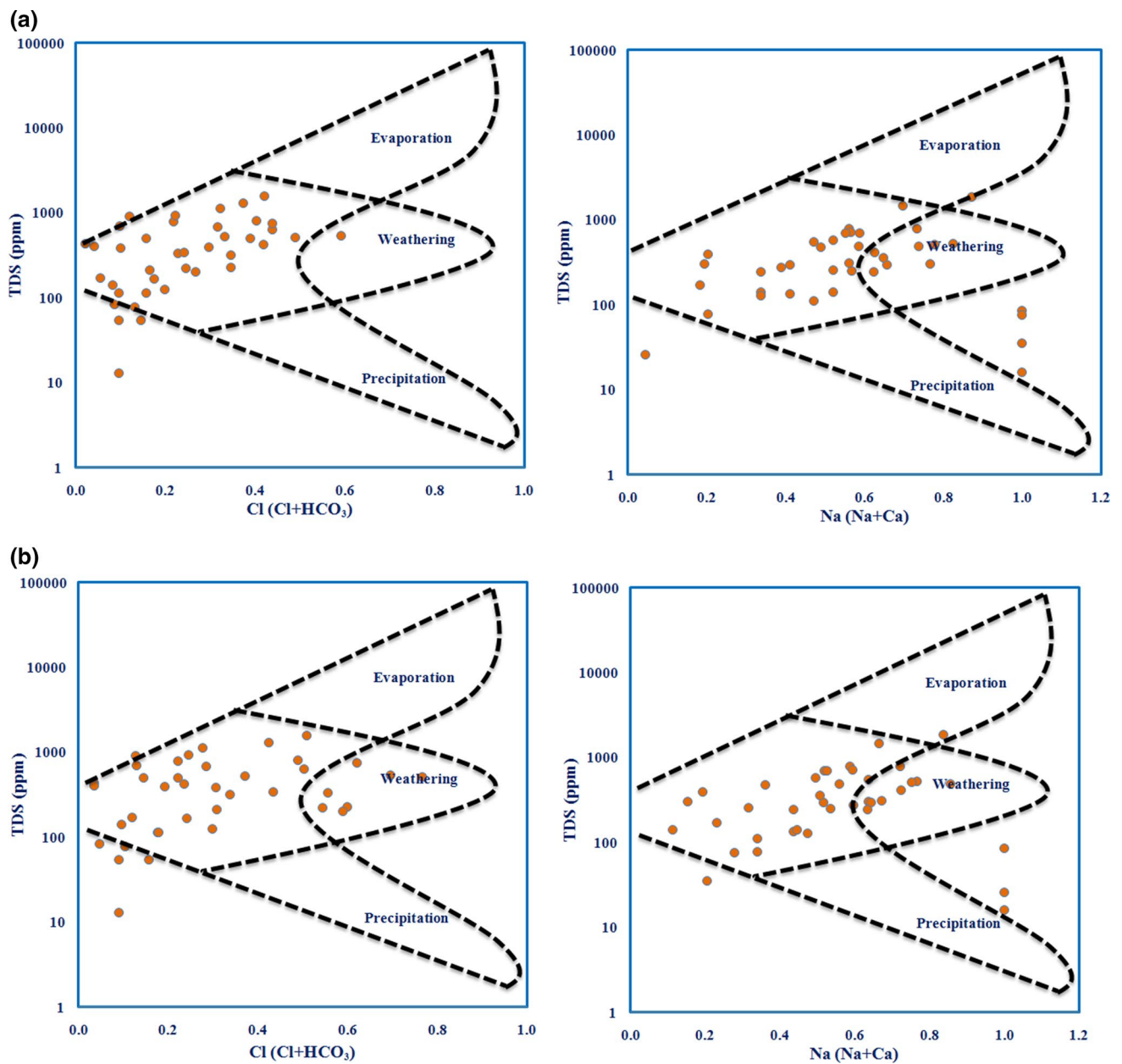


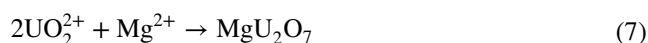
Fig.5 Gibbs plot for water samples from kanniyakumari district **a** pre-monsoon **b** post-monsoon

Table 7 Weight and relative weight of different water quality parameters

Chemical parameter	Weight (w_i)	Relative weight (W_i)	BIS (2012) (S_i)
pH	3	0.083	8.5
TDS (mg/L)	5	0.139	500
Cl (mg/L)	5	0.139	250
Ca ²⁺ (mg/L)	3	0.083	75
Mg ²⁺ (mg/L)	3	0.083	30
SO ₄ ²⁻ (mg/L)	3	0.083	200
NO ₃ ⁻ (mg/L)	4	0.111	45
F ⁻ (mg/L)	5	0.139	1
U (μg/L)	5	0.139	60 (AERB, India)
	$\Sigma w_i = 36$	$\Sigma W_i = 1$	

potential (ORP) values are -57 to 26 mV in pre-monsoon and -78 to 8 mV in post-monsoon. From the ORP values, post-monsoon samples have a higher reducing condition than pre-monsoon. Under the reducing condition, U(VI) is reduced into U(IV), and therefore the aqueous uranium concentration decreases. The previous studies also confirm that under higher oxidizing conditions (68 – 223 mV), more uranium will be dissolved (0.79 – 71.93 μg/L) [52]. The saturation index of uraninite was estimated using the PHREEQC program, shows the average value of -28.8 (pre-monsoon) and -28.7 (post-monsoon) represents UO₂ has under saturated conditions (i.e., less soluble) in the groundwater of the study area (Table 6).

Comparing the correlations of pre-monsoon and post-monsoon parameters suggests the correlation between uranium with PO₄³⁻ and Mg²⁺ decreases during post-monsoon. These parameters' average values also decrease from pre-monsoon (PO₄³⁻- 0.6 ppm; Mg²⁺- 31 ppm) to post-monsoon (PO₄³⁻- 0.3 ppm; Mg²⁺- 26 ppm). It may be due to the formation of complexes of aqueous uranium ion with phosphate and magnesium under these reducing conditions by the following reactions [53, 54] (Eqs. 7–8).

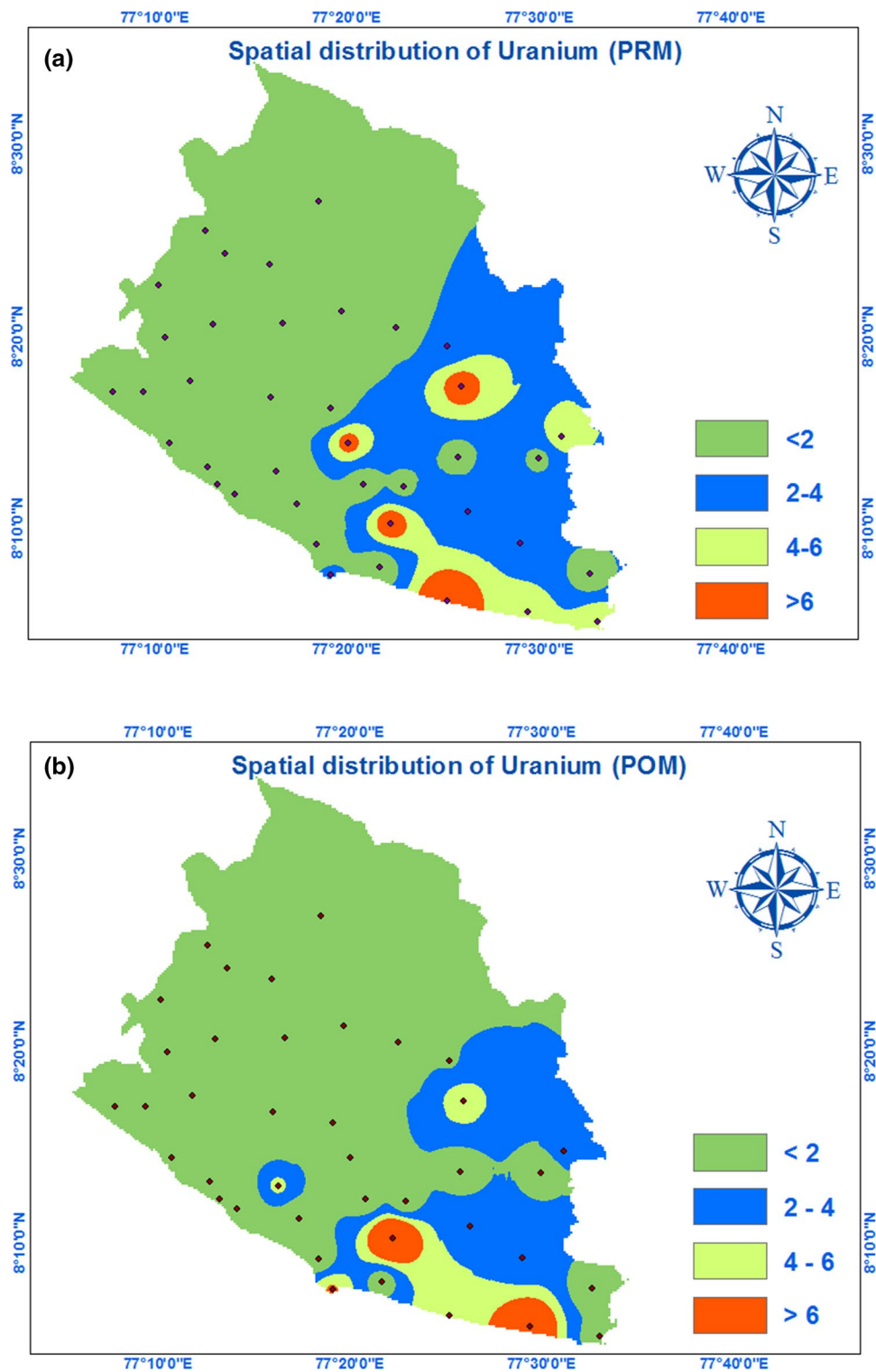


These precipitation reactions may decrease the phosphate and magnesium ions to the study area's uranium ions. Manakudi (S-31), Muttam (S-25), and Karugankuzhi (S-20) have high uranium concentration in the post-monsoon compared to pre-monsoon season, maybe due to the rock-weathering under the favorable geological conditions (i.e. Manakudi and Muttam are placed in peninsular genesis, karugankuzhi from the charnockite geological units), depth and contact time of the water with rocks. Therefore, this district has high background radiation places with less uranium concentration in groundwater because of the groundwater's alkaline and reductive nature in pre- and post-monsoon.

Conclusions

The present study shows that Manavalakurichi (S-13) and Indranagar Junction (S-8) in Kanniyakumari District have high background radiation due to the monazite sand deposits in the coastline. All the collected samples show that the uranium concentration is within the permissible limits of 60 μg/L (AERB) and 30 μg/L (WHO). The present study shows that the samples collected from Puthenthurai (S-30) and Vembanur (S-27) have a maximum uranium concentration compared to the other samples. The present study shows that the dolomite and calcite minerals are in saturated condition and the uraninite minerals have under-saturated conditions. It suggests that the uranium gets dissolved from the autunite and saleeite minerals during pre- and post-monsoon seasons. The leaching of uranium from monazite placer deposits in the Kanniyakumari district is minimized from the pre-monsoon to post-monsoon season due to the alkaline and reductive nature of the groundwater.

Fig.6 Spatial distribution of Uranium (ppb) in Kanniyakumari district **a** pre-monsoon (PRM) **b** post-monsoon (POM)



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