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ORIGINAL ARTICLE

Spatio-temporal Identification of Regions with Anomalous Values of ^{222}Rn in Groundwater of Madurai District, Tamilnadu, India

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Abstract Measurement of dissolved radon (^{222}Rn) activity concentration in groundwater samples from private and public hand pumps, and in bore wells located at Madurai district of Tamilnadu, India, are presented. The study attempts to identify the background value of ^{222}Rn in groundwater of hard rock terrain and the main aquifer contributing ^{222}Rn, and to determine if any correlation exists with observed field parameters. Measured parameters included pH, TDS, Temperature and ^{222}Rn in 42 samples for two seasons (South West Monsoon [SWM] and North East Monsoon [NEM]). The results show that the ^{222}Rn activity concentration of the samples ranged from 0.049 to 59.952 Bq/L in South west monsoon and 0.12 to 211.60 Bq/L in North east monsoon. The higher activity was noted in NEM and the highest ^{222}Rn concentrations were observed in granitic terrains in both seasons. The average values of the parameters studied shows that there is a general decrease of TDS and Temperature, but an increase in ^{222}Rn and pH during NEM. The spatial representation of the activity shows that maximum values are in the north eastern part of the study area. Further, correlations between the measured parameters show that temperature has a negative correlation to the samples of charnockite formation during both seasons; pH and TDS also show negative relationships to ^{222}Rn during SWM.

Keywords Radon (^{222}Rn) · Groundwater · Granitic terrain · Seasonal variation

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1 Introduction

The ^{222}Rn in groundwater is mainly due to the available concentration of the source ^{226}Ra in groundwater. The hard rock aquifers have groundwater accumulated in cracks and interconnected joints; the movement of groundwater is very slow which indirectly enhances the dissolution of the aquifer matrix. The increase of the residence time in hard rock aquifers is generally reflected in the higher Electrical conductivity (EC) (Chidambaram 2000; Srinivasamoorthy et al. 2008). Having a short half-life, ^{222}Rn in groundwater attains a secular equilibrium with ^{226}Ra in the aquifer matrix in the regions with low velocity or with increased residence time. Similar to that of the major ions (e.g., Cl^-, SO_4^{2-}, HCO_3^-, Na^+, Ca^{2+}, Mg^{2+}, etc.), ^{222}Rn can also migrate in groundwater from its source along the direction of movement of water (Przylibski et al. 2004). It has been inferred by earlier researchers that the grade of metamorphism plays a significant role in controlling the activity concentration of ^{222}Rn in groundwater (Butsaert et al. 1981).

In the hard rock aquifers during non-monsoon periods or due to the failure of monsoon, surface water sources area dries up. As a result, groundwater serves as the main source of water for drinking and domestic purposes. In this case, if the available groundwater is rich in ^{222}Rn and its radioactive daughter products, it will create health hazards when entering the human chain. Though ^{222}Rn has a very short half-life, (3.82 days) the time interval between the pumping of groundwater and its consumption is generally well within this half-life period. There have been several studies to understand the impacts of ^{222}Rn on human health. These include lung cancer (Cothern et al. 1986), an increased incidence of childhood leukaemia and possibly gastric cancers (Akerblom 1994) for users of ^{222}Rn-rich groundwater.

The direction and velocity of groundwater movement has been traced by studying ^{222}Rn in regional and local scales (Miller et al. 1990; Cable et al. 1996; Crotwell and Moore 2003; Schwartz 2003; Desideri et al. 2006). Further, ^{222}Rn has also proved to be an indicator of seismic and volcanic activities (King 1986; Virk et al. 1997; Wu et al. 2003). The persistence of openness of fracture can be marked by means of ^{222}Rn results (Wu 2007); moreover, it also helps to determine the degree of fracture openness. According to Mulligan and Charette (2006) the radon-based estimate of SGD (Submarine groundwater discharge) can help us to measure the discharge of fresh groundwater from the coastal aquifers to the sea.

Since no study exists to understand the ^{222}Rn activity concentration in the groundwaters of Madurai region, an attempt has been made in this paper to study the seasonal variation of ^{222}Rn and its distribution in groundwater, and to correlate them with various field parameters.

2 Methods and Materials

2.1 Study Area

The study area is Madurai district located in the southern part of Tamilnadu, which is bounded between Latitude 9°30′.00 and 10°30′.00 N and Longtitude 77°00 and 78°30′E and covers an area of about 3,741 km^2. It is bordered to the east by Sivaganga, to the north by Dindigul and Thiruchirapalli, Theni to the west, and Virudhunagar district to the south (Fig. 1). The study area is predominantly covered by crystalline formations comprising significantly fissile Hornblende Biotite Gneiss followed by Charnockite, Granitic intrusions, Quartzite and flood plain alluvium (Fig. 1). Graphite bearing hornblende gneiss and calc-granulites of the study area are reported to have higher amounts of uranium (12–28 ppm) than schist (Pandey and

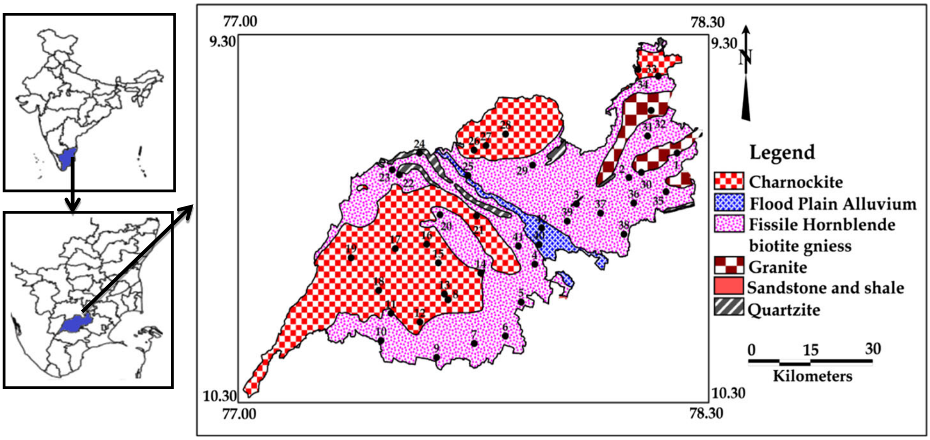

Fig. 1 Lithology and sampling points of the study area (modified after Thivya et al. 2013a, b)

Krishnamurthy 1995). The flood plain alluvium occurs along the Vaigai river course. Groundwater occurs in unconfined conditions in regions with higher weathered thickness, and is also reported to occur in two different fracture zones namely, shallow and deeper zone. In the deeper fracture zone of the study area, groundwater is reported to exist under semi-confined to confined conditions (CGWB 2007). It was observed that more than 30 % of the wells yield between 1 and 3 L/s and 29 % of the wells yield below 1 L/s (CGWB 2007). In general, the groundwater flow is inferred to be from the NW to SE (Thivya 2013). The yield is mainly governed by the distribution of fractures in the rock, or the degree of interconnectivity. Vagai and Mullaiperiyar are the major rivers of the study area. The study of a long-term data on rainfall from 1901 to 2004 shows that north east monsoon represents 47 % and the south west monsoon 32 % of the total rainfall received. The temperature ranges from 15 to 41 °C and the climate is subtropical. The relative humidity during north east monsoon is high and varies from 45 to 85 %.

2.2 Collection of Samples

A total of about 42 samples were collected from hand pumps and borewater representing the entire district for two seasons, i.e., South West Monsoon (SWM) in the month of August and North East Monsoon (NEM) in the month of November. A portable water analysis kit was used to for insitu measurements of pH, temperature and TDS (Thivya et al. 2013a, b). A Thermo Orion 3 star bench top meter was used for measuring pH and temperature. The pH electrode was calibrated by three buffers (pH 4.01, 7.00 and 10.00) to get accurate results. The accuracy of pH and temperature was ±0.002 and ±0.1 °C, respectively. TDS was measured by the Eutech handheld instruments. The accuracy was ±1 % with range of 100, 1,000 ppm and 10.00 ppt. 250 mL water samples were collected separately in glass bottles for ^{222}Rn analysis. The sample was allowed to flow for 10 min before the collection in order to avoid the interference of old water which is already present in the well casing. The atmospheric contact of the water in casing would have resulted in loss of ^{222}Rn by decay or due to outgassing. The ^{222}Rn sampling was carried out carefully by the fact that the gas easily escapes from water. Care was also taken to collect the sample without bubbling. The collected samples were analyzed in the field within 1–3 h of collection; according to the time of analysis, a decay correction was also carried out.

2.3 Radon Activity Concentration Measurements

RAD7 of Durridge Company, USA, was used in determining ^{222}Rn concentration in water. The RAD7's solid state alpha particle detector is almost completely insensitive to beta or gamma radiation, so there was no interference from beta-emitting gases or from gamma radiation fields. The most likely effect of high levels of beta or gamma radiation will probably be an increase in detector leakage current and increased alpha peak width. Typical environmental levels of beta and gamma emitters have absolutely no effect on the RAD7. The overall calibration accuracy of RAD7 is about ±5 %.

The RAD7 (Fig. 2) setup consists of three components: 1. Vial with sample; 2. the desiccant tube; and 3. the Alpha detector. The RAD7 instrument has a convenient stand for the desiccant tube placed between two clasps present in the lid, with provision for the vial in the foam cavity. In principle, it is a closed loop system, where the flow rate is independent of the volume of air and water; but the volume of air and water are constant. The ^{222}Rn extracted from the sprayed water is recirculated as air until a state of equilibrium is developed. The system attains the state of equilibrium when no more ^{222}Rn can be extracted from the water, and this process persists for 5 min. More than 95 % of available ^{222}Rn in water is removed before the state of equilibrium (Singh et al. 2010).

3 Results and Discussion

3.1 Quality and Status

There is a seasonal variation in higher concentration levels of ^{222}Rn. In SWM, the highest value is observed as 60 Bq/L, whereas in NEM the value is 211.6 Bq/L at the same sampling location (Karungalakudi) in the study area (Table 1). The USEPA permissible limit of ^{222}Rn in drinking water is 11 Bq/L (USEPA 2003). In the study area, 16 % of the samples exceeded the

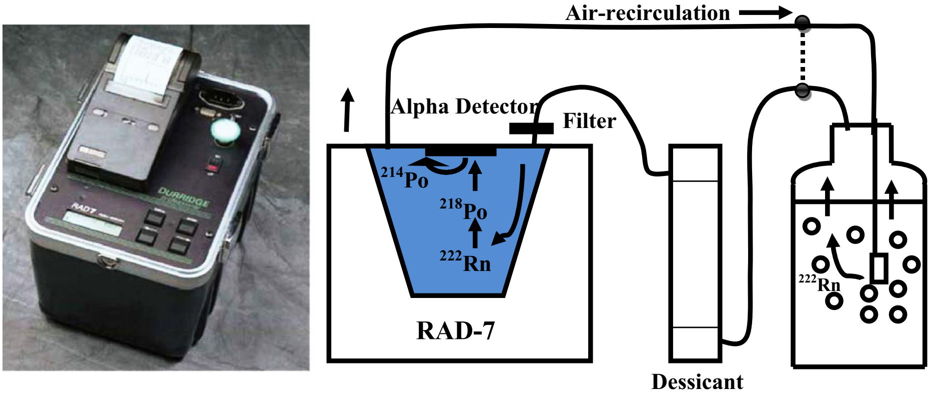

Fig. 2 Radon meter (RAD7) and Schematic diagram of the measurement system, the dotted line is for a bypass loop which is used after air- water equilibration is reached (after Singh et al. 2010)

Table 1 The value of ^{222}Rn, pH, TDS and temperature (Temp) for the sample of both seasons with measurement accuracy of ^{222}Rn for each sample

Sample no.	Location name	SWM (South west monsoon)				NEM(North east monsoon)			
		^{222}Rn (Bq/L)	pH	TDS (mg/L)	Temp (°C)	^{222}Rn (Bq/L)	pH	TDS (mg/L)	Temp (°C)
1	Keelavalavu	7.6±0.3	7.1	1,078	30.4	7.3±0.29	7.2	1,371	30.8
2	Chinnasooragundu	23.6±1.1	7	499	29.8	17±0.83	7.4	362	30.6
3	Othakadai	36.9±1.2	6.5	96.4	31.9	91.9±4.4	6.9	78.9	31.7
4	Perungudi	5.9±0.2	6.9	530	32.9	8±0.35	6.9	491	32.5
5	Koodakovil	1.3±0.05	6.9	1,011	33.5	0.5±0.01	6.9	749	29.6
6	Nochikulam	2.0±0.07	7.1	756	35.3	2.7±0.11	7.2	482	30.3
7	Kallikudi	2.1±0.1	6.7	1,081	34.3	7.7±0.35	6.9	1,056	31
8	Villur	9.9±0.48	6.8	1,067	32.3	1.6±0.06	7	573	31.4
9	Pudhupatti	1.5±0.06	7.4	980	34.6	0.2±0.01	8.5	1,044	31.8
10	Koovalapuram	2.9±0.12	7.1	306	32	1.9±0.07	7.5	258	30.9
11	Peraiyur	3.5±0.15	6.9	639	34	7.6±0.34	6.9	684	29.9
12	T.kallupatti	1.7±0.07	7.1	1,058	35.8	4.7±0.21	7.1	1,320	31.3
13	Tottiapatti	1.6±0.07	6.7	2,037	35.7	3.3±0.14	7.2	2,031	31.1
14	Tirumangalam	1.8±0.06	7.2	1,154	33.8	1.5±0.05	7.4	771	31.2
15	Ammapettai	1.2±0.03	7.4	686	33.5	0.3±0.01	7.3	667	30.7
16	Sivankoilpatti	0.2±0.005	7.8	487	32.6	0.3±0.006	7	386	31.6
17	Perumalkoilpatti	0.1±0.003	7.5	1,089	36.5	0.2±0.004	7.3	790	31.7
18	Athipatti	4.0±0.19	7.5	1,175	31.8	4.2±0.20	7.6	918	31
19	Elumalai	0.7±0.03	7.1	1,066	31.1	4.2±0.19	6.9	1,044	30.6
20	Chellampatti	1.9±0.10	7.3	729	27.9	2.2±0.10	7.1	678	30.6
21	Checkanurani	4.3±0.19	6.7	1,160	26.5	2.6±0.11	6.7	1,046	31
22	Kulathupatti	10.9±0.48	6.6	306	32.5	4.5±0.20	7.4	250	32
23	Uthappanaickanur	1.0±0.26	7	910	31.7	1.4±0.04	6.7	1,146	30.7
24	Kalyanipatty	7.1±0.32	7.2	658	32	0.2±0.01	7.1	632	27.7
25	Sholavandan	2.0±0.07	7.1	893	32.5	1.2±0.03	7.1	902	30.1
26	Vadipatti	9.5±0.43	7.3	629	31.9	20.3±0.35	7.1	1,067	31.3
27	Kachaikatti	6.0±0.27	7.3	311	32.5	10±0.72	6.9	1,068	30.7
28	Mettupatti	5.7±0.25	7	473	31.2	7.1±0.30	6.6	740	31.5
29	Alanganallur	6.7±0.31	7.5	686	31	15.4±0.76	7.3	526	30
30	Melur	9.2±0.41	6.8	1,185	33.2	5.4±0.23	6.9	1,314	30.6
31	Near Kallampatti	0.8±0.02	7.9	1,177	32.3	3.8±0.16	7.4	1,124	30.4
32	Karungalakudi	60.0±2.7	6.9	1,059	31.1	211.6±9.5	7	1,060	30.7
33	Kottampatti	17.4±0.66	7	416	30	10.3±0.48	7.3	372	29
34	Thanichiyam	24.3±1.1	6.8	462	31.3	6.9±0.29	7.6	467	30.6
35	Othapatti	3.3±0.14	6.8	720	29.8	3.2±0.14	7	643	31.1
36	Near Thiruvadhavoor	11.3±0.54	5.9	770	32	0.8±0.02	7.5	637	32.7
37	Panaikulam	5.5±0.26	7.3	1,076	29.4	1.8±0.06	7.5	1,415	32.6
38	Poonjuthi	16.4±0.81	7.2	443	26.7	15.5±0.71	7.4	406	31.7
39	Karupayurani	3.6±0.17	6.4	2,049	29.7	2.3±0.09	6.6	2,068	30.1
40	Avaniyapuram	0.9±0.03	5.2	3,081	30.8	2.2±0.07	6.2	3,050	31

Table 1 (continued)

Sample no.	Location name	SWM (South west monsoon)				NEM(North east monsoon)			
		^{222}Rn (Bq/L)	pH	TDS (mg/L)	Temp (°C)	^{222}Rn (Bq/L)	pH	TDS (mg/L)	Temp (°C)
41	Thiruparakundram	9.8±0.46	6.1	739	30	5.9±0.26	7	568	29.8
42	Meenakshiamman temple	5.5±0.25	8	1,017	30.5	6.2±0.28	7.7	816	28

permissible value in SWM and 11 % in NEM. In SWM, 7 samples (Chinnasooragundu, Othakadai, Karngalakudi, Kottampatti, Thanichiyam, near Thiruvadhavoor and Poonjuthi) were above the permissible limit, whereas in NEM 6 samples (Chinnasooragundu, Othakadai, Vadipatti, Alanganallur, Karungalakudi and Poonjuthi) were above the permissible limit.

Several attempts have been made in India to study ^{222}Rn concentration in groundwater (Table 2). The various studies show that highest ^{222}Rn concentration is observed in Budhakedar, Gharwal Himalaya (Prasad et al. 2009). The study area Madurai presents the second highest ^{222}Rn concentration up to 211.6 Bq/L (Table 2). The concentrations of ^{222}Rn in the regions like Himachal Pradesh, Uttarkasi, Doon valley and Siwaliks pertain to the tectonically active sedimentary terrain. Furthermore, those present in Punjab are related to higher uranium concentration in groundwater. In the present study, the ^{222}Rn concentration pertains to hard rock aquifer with higher uranium and with good fracture intensity (Thivya 2013). The study also identified that a maximum uranium value of 157 μg/L was observed in this region. ^{222}Rn, in any form, either in water or rock, can accumulate in open fractures. CGWB (2007) observations showed that there are two prominent fracture zones: 1. Less than 50 m; and 2. More than 100 m. In each of these zones, 2–3 fracture forms were identified (CGWB 2007).

Figure 3 presents the spatial distribution of ^{222}Rn in groundwater during SWM, which shows that 2,442 km^2 of the area has concentrations below 0.69 Bq/L. The higher ^{222}Rn

Table 2 A comparison of ^{222}Rn in groundwater in different parts of India with the present study area

S.No	Area	^{222}Rn level (Bq/L)		Reference
		Min	Max	
1.	Budhakedar, Garhwal Himalaya	8	3,047	Prasad et al. (2009)
2.	Bathinda and Gurdaspur districts of Punjab	3.0	8.8	Virk et al. (2001)
3.	Doon Valley of the Outer Himalaya.	25	92	Choubey et al. (2003)
4.	Varahi and Markandeya river basins, Karnataka State, India	0.2 2.21	10.1 27.3	Somashekar and Ravikumar (2010)
5.	Uttarkashi	0.00051	89	Ramola et al. (2008)
6.	Upper Siwaliks of Kala Amb, Nahan and Morni Hills of Haryana and Himachal Pradesh states	0.87	32.10	Singh et al. (2008)
7.	Madurai, Tamilnadu	0.1 0.1	60.0(SWM) 211.6(NEM)	Present study

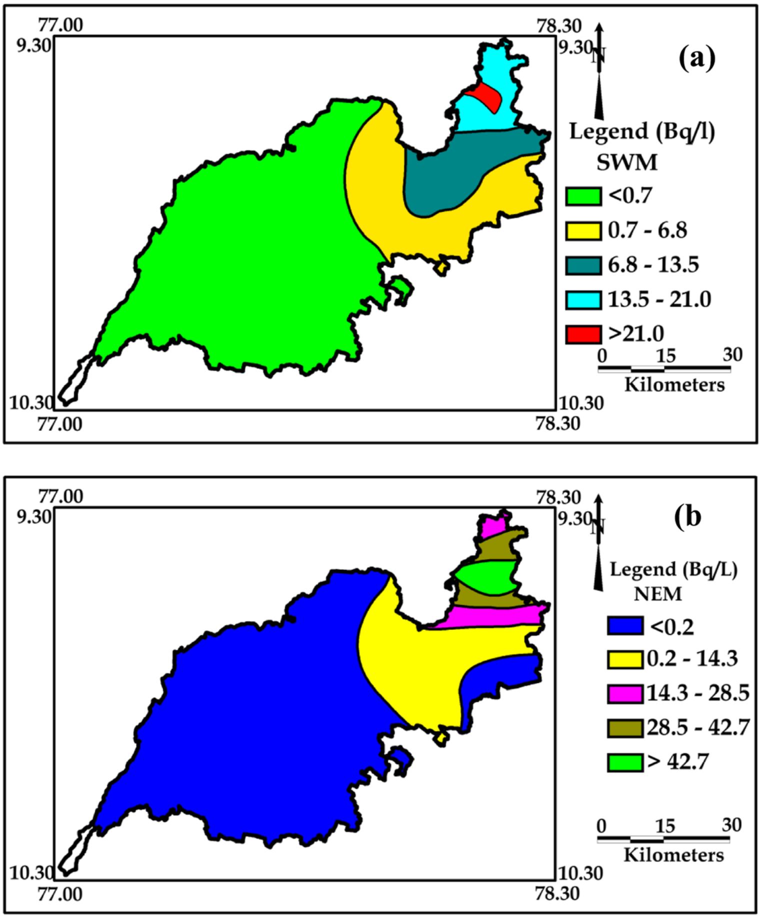

Fig. 3 Spatial distribution of ^{222}Rn in (**a**) SWM and (**b**) NEM

activity concentrations above 21 Bq/L, extends in an area of about 28 km^2 in the north eastern part of the study area. Concentrations between 0.70 Bq/L and 20.06 Bq/L extend for an area of about 1,245 km^2. It is believed that the geology is the most important factor controlling the source and distribution of ^{222}Rn, because it is controlled by the chemical composition of the rocks and soils from which ^{222}Rn originates. Relatively high levels of ^{222}Rn are associated with granites, phosphatic rocks, ironstones and aluminum shales (Appleton 2008). In some granites, most uranium is found as uraninite, which is relatively easily weathered to radium from which ^{222}Rn can escape into groundwater (Appleton 2008).

Concentration values lower than 0.2 Bq/L were found in about 2,712 km^2 during NEM in the study area. The spatial representation of highest ^{222}Rn concentrations above 42.8 Bq/L covers an area of about 89 km^2 in the north eastern part of the study area. The concentration between 0.3 and 42.7 Bq/L covers an area of about 916 km^2 (Fig. 3). There is a gradual increase of ^{222}Rn activity concentration along the north eastern part of the study area which is represented by pink granitic intrusions.

Hence it is inferred that regions with elevated levels of ^{222}Rn are represented in the granitic terrains in both seasons. Water travelling through granitic aquifers was found to have higher levels of ^{222}Rn, uranium and thorium series of radionuclides (Otwoma and Mustapha 1998). The samples were collected through hand pumps and it is believed that the ^{222}Rn contents in hand pumps have a sensible relationship to geological and geohydrological patterns (Prasad et al. 2009). The transport of groundwater is enhanced during NEM due to the recharge after the rainfall of this or previous monsoon. The dissolution and chemical reaction favoring the release of major ions and ^{222}Rn is enhanced immediately after the monsoon (Prasad et al. 2008). The study was carried out during the monsoon period and it has to be noted that in India SWM follows summer and NEM follows SWM. Hence, the precipitation of salt along the available pore space is more prominent during summer and the dissolution starts during SWM and subsequently the gas gets enhanced during NEM.

The lithology wise characterization of the ^{222}Rn distribution in groundwater shows that higher concentration was observed in the granitic terrain followed by quartzite in both seasons (Table 3). The order of dominance of ^{222}Rn in groundwater samples representing both seasons are as follows: granite > quartzite > charnockite > hornblende biotite gneiss > flood plain alluvium.

Groundwater from quartzites shows higher concentrations of ^{222}Rn next to that of granitic aquifer. It is known that quartzites have many small fractures and cracks (Kumar et al. 2014). The nearby granites serve as the source of ^{222}Rn to quarzites, as they occur along the movement of water. ^{222}Rn gets enhanced as it passes through nano-pores (Rama and Moore 1984). ^{222}Rn from the rocks through the process of alpha recoil accumulates by dissolution in the interstitial water. Furthermore, these gases travel rapidly through the microfractures to larger fractures and ultimately reach the sampling holes. The activity of ^{222}Rn can be higher in groundwater systems when the fracture width or aperture becomes smaller. Since the formation does not have huge interconnected pore spaces, it does not serve as a good aquifer (Kumar et al. 2014) and hence has a lesser volume of water. It is also inferred that the volume of water present in the aquifer is inversely proportional to the concentration of ^{222}Rn in the formation (Butsaert et al. 1981). The diffusion rate of ^{222}Rn varied according to the nature of fracture. In microfractures, the activity of ^{222}Rn is higher than in larger fractures. But the openness of the interconnected fractures can be identified only if the ^{222}Rn level attains its detection limit when sampled, in hard rock terrains.

The review of ^{222}Rn concentrations in groundwater from granitic terrains show higher values (Table 4). The observably enhanced concentrations of ^{222}Rn in waters of granitic aquifer are due to the moving out of ^{222}Rn gas from the parent rock and dissolving in the surrounding water under geologic pressure; it then tends to get released into the atmosphere at normal atmospheric conditions (Somashekar and Ravikumar 2010). In general, it is documented that larger concentrations of ^{222}Rn are typically associated with granitic rocks that contain elevated concentrations of ^{238}U (typically ten or more ppm). Hall et al. (1987) measured ^{222}Rn concentrations in waters from the granitic regions and inferred to have excess

Table 3 Lithology wise ^{222}Rn concentration in Madurai

Lithology	South West Monsoon		North East Monsoon	
	Min(Bq/L)	Max(Bq/L)	Min(Bq/L)	Max(Bq/L)
Fissile hornblende biotite gneiss	1.0	16.4	0.2	15.5
Charnockite	0.1	24.3	0.1	20.3
Quartzite	7.1	36.9	0.2	91.9
Flood plain alluvium	0.9	5.5	1.2	6.2
Granite	7.6	60.0	5.4	211.6

Table 4 Few studies on ^{222}Rn distribution in different granitic terrain of the world

S.No	Terrain	Area	^{222}Rn level (Bq/L)		Reference
			Max	Min	
1.	Uranium-rich granite	Sweden	8900	300	Akerblom et al. (2005)
2.	Granitic terrain	Migdonia valley	161	8	Zouridakis et al. (2002)
3.	Granites, amphibolites and migmatites	Helsinki, Finland	32,560	3.7	Asikainen and Kahlos (1979)
4.	Granitic and Precambrian igneous bedrock	Extremadura	1168	0.24	Lopez and Sanchez (2008)
5.	Piedmont region underlain by granites	North Carolina	218	1.7	Loomis (1987)

values (3.7 to 104 Bq/L) than that of other formations. Larger than average ^{222}Rn have also been measured in other ^{238}U naturally enriched sites, such as phosphate bearing rocks. Uranium in the hard rock is also reported in the study area which is mainly hosted by magnetite and allanite occurring as independent grains with flaky graphite and also with inclusions of quartz (Pandey and Krishnamurthy 1995).

3.2 Correlation

To obtain an overall interrelationship between the various sample parameters, correlation analysis was attempted between different field parameters, i.e., ^{222}Rn, pH, TDS and

Table 5 correlation matrix of SWM and NEM

SWM		^{222}Rn	pH	TDS	Temp
^{222}Rn	Pearson Correlation	1			
	Significant level				
pH	Pearson Correlation	−0.173	1		
	Significant level	0.273			
TDS	Pearson Correlation	−0.266	-.397**	1	
	Significant level	0.089	0.009		
Temp	Pearson Correlation	0.247	0.022	-.337*	1
	Significant level	0.115	0.89	0.029	
NEM		^{222}Rn	pH	TDS	Temp
^{222}Rn	Pearson Correlation	1			
	Significant level				
pH	Pearson Correlation	−0.095	1		
	Significant level	0.548			
TDS	Pearson Correlation	−0.067	-.367*	1	
	Significant level	0.674	0.017		
Temp	Pearson Correlation	−0.124	−0.004	−0.11	1
	Significant level	0.435	0.982	0.49	

**Correlation is significant at the 0.01 level (2-tailed)

*Correlation is significant at the 0.05 level (2-tailed) temperature. In SWM, the ^{222}Rn was negatively correlated with temperature, pH and TDS. In NEM, the ^{222}Rn showed a poorly positive correlation with temperature and a negative correlation with the pH and TDS (Table 5). It is also inferred that ^{222}Rn emanation and the rock waters interaction processes are favored by high temperatures. The ^{222}Rn concentration was found to be negatively correlated (−0.70) with the water temperature in few places in India (Prasad et al. 2009), but in the present study the relationship is site-specific depending on depth

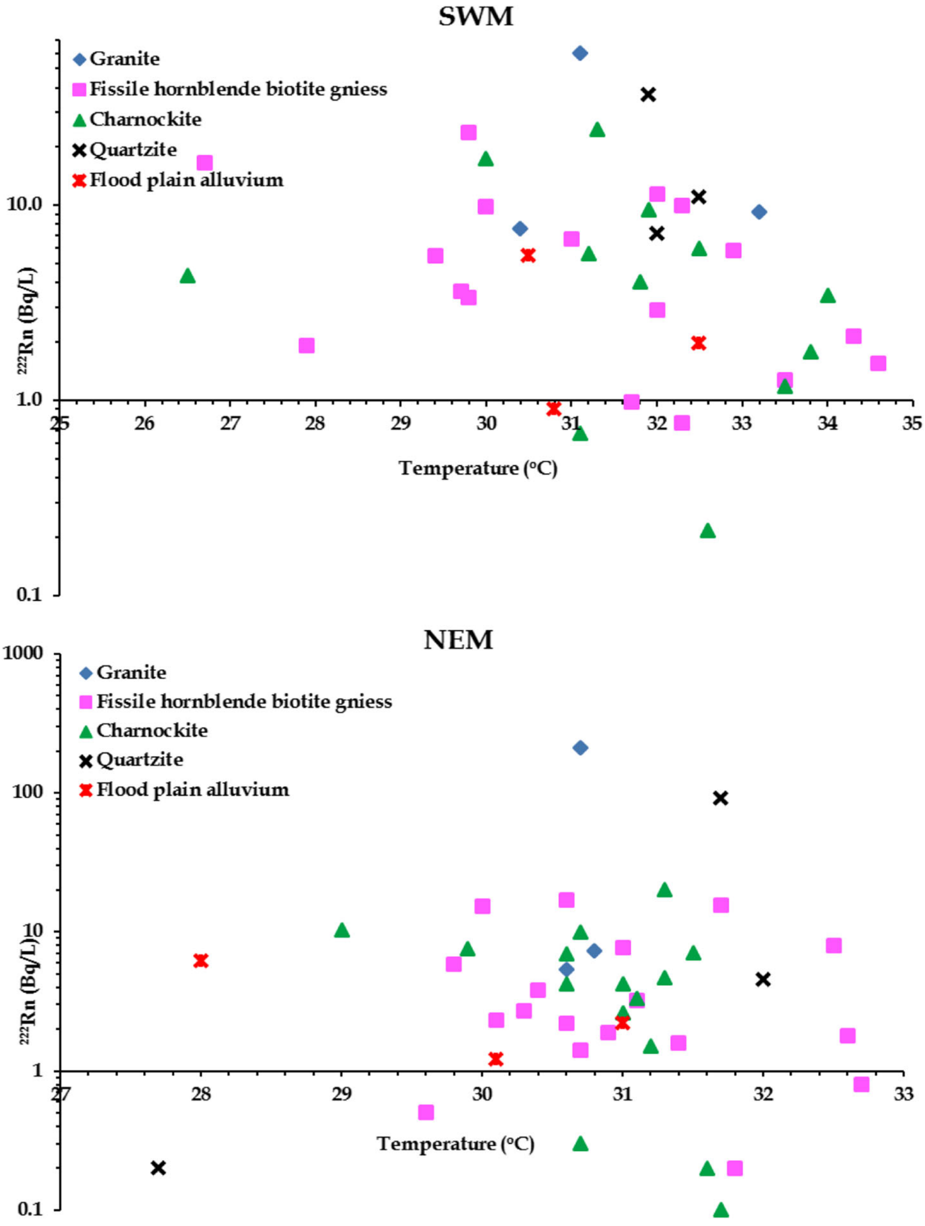

Fig. 4 Bivariate plot of temperature and ^{222}Rn in SWM and NEM

of fracture and the type of the aquifer. In general, the temperature study reveals the fact that it is not a controlling factor for the release of ^{222}Rn.

In SWM, the maximum and minimum temperature was observed in Charnockite rock type (26.5 °C and 36.5 °C, respectively). The lowest ^{222}Rn concentration was related to the highest temperature in SWM. In NEM, the maximum value was observed in fissile hornblende biotite gneissic rocks, whereas the minimum concentration was observed in quartzite. The lower concentration of ^{222}Rn in NEM occurred at the temperature of 31.7 °C. The highest concentration of ^{222}Rn in SWM occurred at the temperature of 31.1 °C, whereas in NEM the

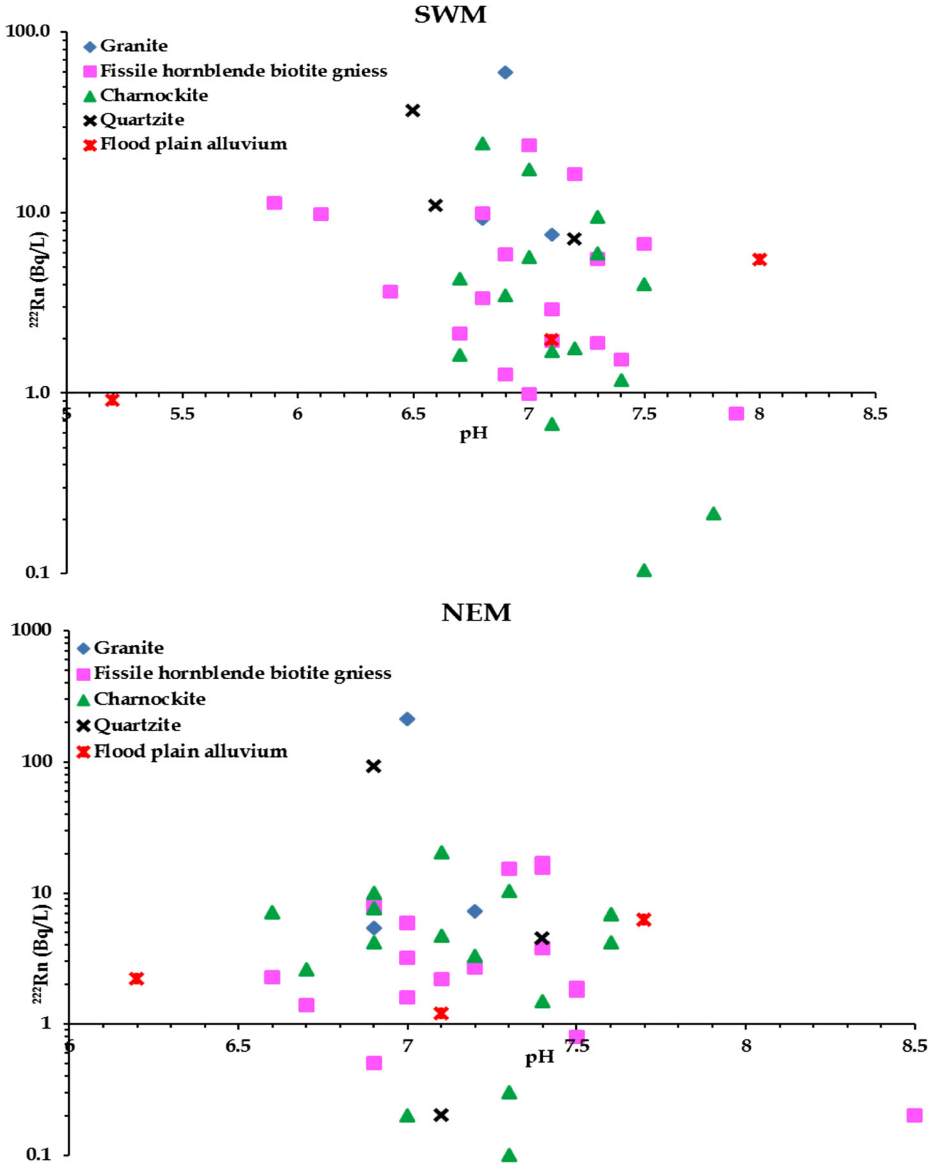

Fig. 5 Bivariate plot of pH and ^{222}Rn in SWM and NEM

respective value was at 30.7 °C (Fig. 4). When the data are studied in total in the correlation matrix, ^{222}Rn content in groundwater has no significant relation to temperature, but when represented graphically with respect to lithology, it can be clearly seen that the samples of

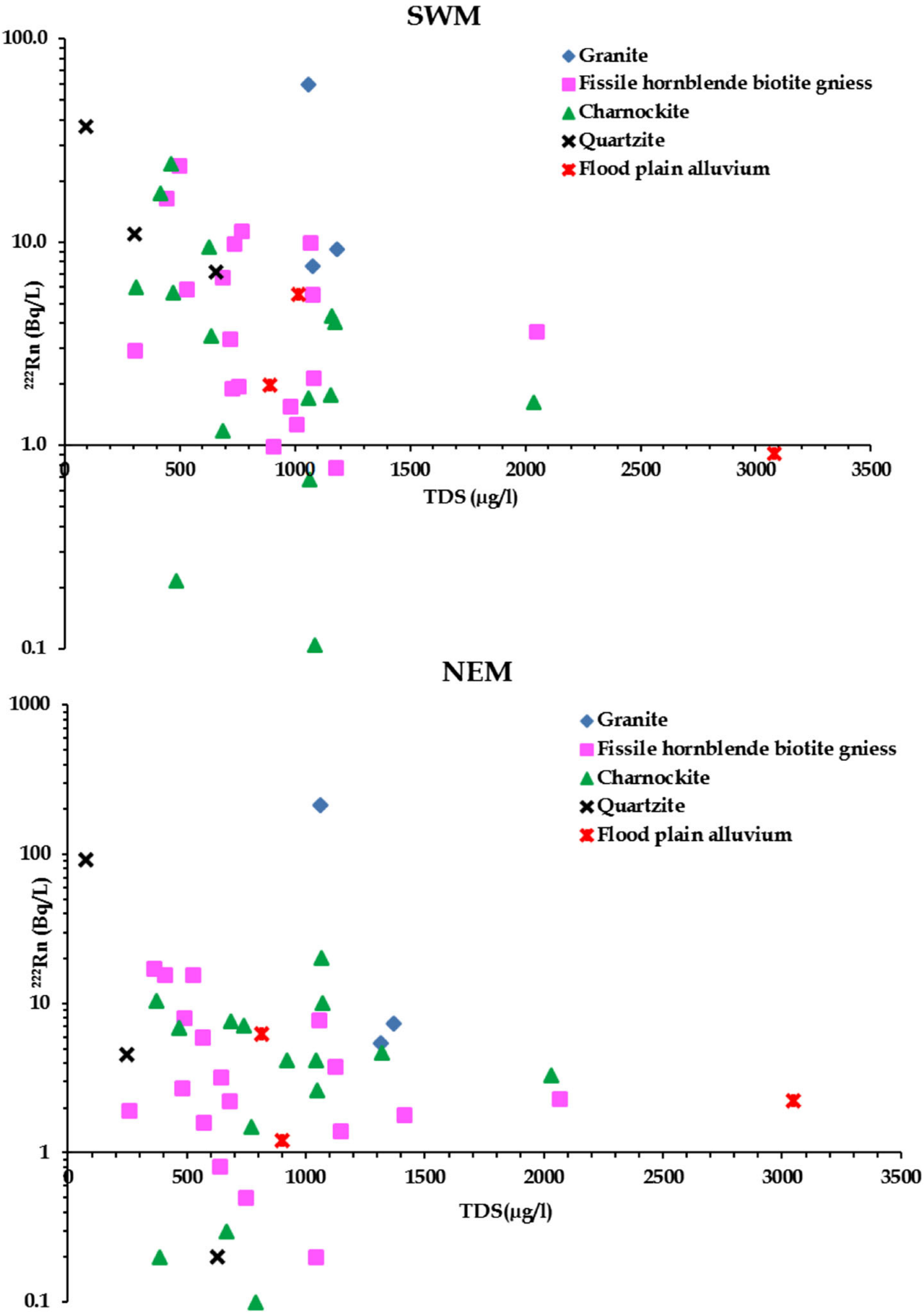

Fig. 6 Bivariate plot of TDS and ^{222}Rn in SWM and NEM

Table 6 R^2 value of ^{222}Rn (Bq/L) with temperature (°C), pH and TDS (μg/L)

Litho units	SWM (Temp)	NEM (Temp)	SWM (pH)	NEM (pH)	SWM (TDS)	NEM (TDS)
Quartzite	0.283	0.232	0.488	0.604	0.725	0.586
Fissile hornblende biotite gniess	0.212	0.009	0.06	0.00005	0.135	0.149
Charnockite	0.165	0.08	0.144	0.015	0.216	0.001
Granite	0.065	0.00006	0.046	0.032	0.353	0.967
Flood plain alluvium	0.181	−0.781	0.766	0.456	0.411	0.129

charnockite aquifer have a negative correlation to the ^{222}Rn concentration. The residence time of the water play a significant role in determining the temperature (Karmegam et al. 2010) in the Charnockite formation. The figure shows that there is no definite correlation of temperature to the concentration of ^{222}Rn in other formation, which indicates that in certain aquifers and site-specific location this relationship persists.

In SWM, the highest and lowest pH values were observed in floodplain alluvium. In NEM, the highest pH value was observed in fissile hornblende biotite gneiss and the lowest value in flood plain alluvium. In SWM, the highest ^{222}Rn concentration occurred at pH value of 6.9 and the lowest at pH value of 7.5 (Fig. 5). In NEM, the highest ^{222}Rn concentration was related to a pH value of 7.0 whereas the lowest to a pH value of 7.3.

The highest concentration of TDS was observed in floodplain alluvium in both seasons. The lowest concentration of TDS was observed in quartzite, in both SWM and NEM. The lowest ^{222}Rn concentration was related to a TDS value of 1,089 mg/L in SWM, whereas, the highest observed TDS value was 1,059 mg/L (Fig. 6). In NEM, the lowest ^{222}Rn concentration had a TDS value of 790 mg/L and the highest ^{222}Rn concentration had a TDS value of 1,060 mg/L.

The correlation was carried out using all samples, which did not provide a clear picture, and hence, an attempt was made to correlate the ^{222}Rn values of the samples to lithology, temperature, TDS and pH. The temperature showed lower R^2 values, indicating that the ^{222}Rn has no or lesser relationship to temperature in certain lithologies of the study area. The R^2 value of ^{222}Rn and pH showed that good correlation exist between these parameters for the samples collected in quartzite and sedimentary formation (floodplain alluvium). Similarly, good correlation of TDS was observed with ^{222}Rn in the quartzite formation in both seasons (Table 6). Good correlation between ^{222}Rn in granite formation and TDS was observed in NEM season. Though the occurrence of ^{222}Rn was reported to be higher in the granitic formation of the study area and the relation between the occurrences of this isotope to observed field parameter shows that ^{222}Rn collected from the samples of quartzitic terrain had good correlation with pH and temperature.

4 Conclusions

The study on the ^{222}Rn activity concentration in groundwater of this region shows that there is a seasonal variation in higher concentration levels in both seasons. 14 % of samples during NEM have higher value and they are noted in the granitic formations of the study area. These higher values are represented in NE part of the study area. The order of dominance of ^{222}Rn in groundwater is as follows granite > quartzite > charnockite > hornblende biotite gneiss > floodplain alluvium. The higher value in granite is mainly due to the presence of uranium bearing minerals. The source of ^{222}Rn in quartzite is mainly due to the ^{222}Rn enriched groundwater flowing from the granitic terrain, as quartzite formation is along the groundwater flow direction. But since it moves through microfractures and the volume of water is lesser it shows an enhanced ^{222}Rn by representing the second highest value in this aquifer of the study area. It shows less correlation with other observed parameters like TDS, pH and temperature, and negative correlation is exhibited in few samples of charnockites. The seasonal variation may be due to the source, fracture systems, and leaching processes occurring in the granite bed rock where leaching reaches maximum during NEM. The consequences of these processes lead to the high ^{222}Rn concentrations in water extracted from the bedrock. The study on the cations and anions, along with the isotope concentrations can help in identifying the complete processes behind the variations of higher ^{222}Rn concentrations.

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