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Health Risk Implication and Spatial Distribution of Radon in Groundwater Along the Lithological Contact in South India

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Abstract

The presence of radioactive elements in groundwater results in high health risks on surrounding populations. Hence, a study was conducted in central Tamil Nadu, South India, to measure the radon levels in groundwater and determine the associated health risk. The study was conducted along the lithological contact of hard rock and sedimentary formation. The concentrations of uranium (U) varied from 0.28 to 84.65 µg/L, and the radioactivity of radon (Rn) varied from 258 to 7072 Bq/m^3 in the collected groundwater samples. The spatial distribution of Rn in the study area showed that higher values were identified along the central and northern regions of the study area. The data also indicate that granitic and gneissic rocks are the major contributors to Rn in groundwater through U-enriched lithological zones. The radon levels in all samples were below the maximum concentration level, prescribed by Environmental Protection Agency. The effective dose levels for ingestion and inhalation were calculated according to parameters introduced by UNSCEAR and were found to be lesser (0.235–6.453 µSvy^{-1}) than the recommended limit. Hence, the regional groundwater in the study area does not pose any health risks to consumers. The spatial distribution of Rn's effective dose level indicates the higher values were mainly in the central and northern portion of the study area consist of gneissic, quarzitic, and granitic rocks. The present study showed that Rn concentrations in groundwater depend on the lithology, structural attributes, the existence of uranium minerals in rocks, and the redox conditions. The results of this study provide information on the spatial distribution of Rn in the groundwater and its potential health risk in central Tamil Nadu, India. It is anticipated that these data will help policymakers to develop plans for management of drinking water resources in the region.

Introduction

Groundwater is a crucial and significant water resource on the global scale (Subba Rao et al. 2020). Overpopulation and superior living standards lead to a rapid demand and increased usage of groundwater during the past few decades (Ji et al. 2020; Li and Qian 2018; Su et al. 2020). Exposures to radioactive elements is one of the major water-quality concerns that have not been investigated extensively. Many studies have stated high U and Rn levels in groundwater of various granitic terrains (Lahermo et al. 1990; Cho et al. 2007, 2015, 2019; Prat et al. 2009; Thivya et al. 2014, 2016a, b; Yun et al. 2017). High Rn levels are commonly observed in granitic terrains that generally contain more U minerals in the rock matrix (Yun et al. 2017). The Rn level in groundwater usually rises with an increase in U content of the soil and bedrock (Knutsson and Olofsson 2002). However, depending on aquifer characteristics, geology, and hydrochemical parameters, radon in groundwater is highly

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variable (Choo and Choo 2019). High levels of Rn can pose health risks to residents and this radioactive element can be transported into the aquifer due to its water-soluble characteristics. The major pathway for Rn migration from source rock is physical transport as a solute in groundwater (Durrance 1986; Nazarof 1992; Waseem et al. 2015). Groundwater systems acquire Rn from aquifer formations predominantly through alpha recoil of Rn as well as diffusion and transfer through mineral grains and fracture networks in a rock mass (Andrews and Wood 1972; Durrance 1986). Rn produces short-lived alpha decay products which may cause health risk to human body if inhaled or ingested (NRC 1999). Drinking water with an exceptionally high Rn content greatly raises the risk of stomach and gastrointestinal cancers (Zhuo et al. 2001; Kendall and Smith 2002; Kendall et al. 2015). However, inhaled radon poses a higher risk than ingested radon (Folger et al. 1994; Khan et al. 2010); 89% of radon-related deaths are attributed to radon inhalation (lung cancers) and 11% due to drinking water (stomach cancers) ingested by radon (USEPA 1999). Identifying the amount of groundwater Rn in household environments is essential to prevent excessive radiation exposure and to quantify potential health risks (Brunskill and Wilkinson 1987; Council 1999; Segovia et al. 2007). Radon migrates through groundwater into households and other structures, generating a health hazard (Badhan et al. 2010). Dissolved radon is released into the indoor atmosphere when water is used for bathing, washing, and other domestic uses.

Groundwater from granitic aquifers contains higher Rn levels greater than 100,000 pCi/L (Asikainen and Kahlos 1979; Brutsaert et al. 1981; Snihs 1973). It was reported that Rn levels in groundwater from sedimentary aquifers were less than 500 pCi/L (Andrews and Wood 1972; Gorgoni et al. 1982; King et al. 1982). Besides U content of source, the Rn level in groundwater also is affected by other geological and hydrological conditions, such as dispersal of nuclides, groundwater flow, and hydrogeochemistry (Nandakumaran et al. 2015).

In previous studies in the study area conducted by Thivya et al. (2014, 2015, 2016a, b, 2017), these authors reported that higher Rn concentrations in groundwater were associated with higher U concentration in granitic rocks. In these studies, Rn values varied from 0.20 to 211 Bq/L, but no attempt was made to calculate the annual radon dose to humans through drinking water. Adithya et al. (2016, 2020) also studied the overall water quality and U geochemistry of the study area and identified that rock weathering was the major process in the granitic terrain that played a key role in U release into groundwater. This mechanism was believed to be facilitated by variations in the Oxidation Reduction Potential (ORP) of the water.

These previous studies mainly concentrated on describing the general water chemistry and U concentrations in groundwater in this water-scarce region. Although Rn levels were reported in the previous studies, several aspects were not addressed, including the human health risks calculated using estimates of the average annual dose, detailed lithological data, land use controls, and the relationship between U and Rn in the groundwater. Hence, the present study focuses on describing: (1) the geochemical controls of Rn in groundwater, (2) the qualitative analysis of Rn related to the lithology and landuse evaluated using a GIS model, (3) health risk assessment with respect to exposure to Rn calculated from estimates of the effective dose.

Materials and Methods

Study Area

The study area is located in the central part of Tamilnadu (South India), which comprises the districts of Madurai, Dindigul, Trichy, Pudukottai, and Sivaganga (Fig. 1). It covers a total surface area of 4311 km^2 and is limited to East 09°53′24″–10°20′60″ latitudes and North 78°1′48″–78°48′36″ longitudes. Vaigai River is the major seasonal river, which originates from the western Ghats. The average precipitation is 950 mm/yr. The study area has a diverse geological terrain consisting of hard rock, Cenozoic sedimentary rocks with conglomerate beds, and granite intrusions. The geological settings of the region have been reported favourable for likely uranium deposit (Thivya et al. 2016a, b, Adithya et al. 2019). The major rock types present in the central, northern, western, and southern portions of the study area are fissile hornblende biotite gneiss (FHBG), Charnokites on the west, followed by the Hornblende biotite gneiss (HBG) on the NE (Fig. 2). In the study area, there are six categories of land use patterns, including agricultural land, water bodies, tank, wasteland, forest, and land build-up area (Fig. 3a). The western portion is covered by the forest, and the agricultural land is spread throughout the study area. Wasteland is predominantly observed in the eastern side of the study area. Weathered fractured crystalline formation and porous formation are the two major aquifers in the study region. The aquifer system presents in both hard and sedimentary formations, including unconfined and confined condition. In the hard rock terrain, groundwater occurs under semiconfined to confined condition and confined condition in sedimentary terrain. The maximum yields in sedimentary and hard rock aquifers are 12% and 1.5%, respectively, and the transmissivity varies between 1 and 5 m^2/day in shallow aquifers and 1–25 m^2/day in deeper aquifers. The storativity varies from 7.5×10^{-5} to 3.59×10^{-4} in the sedimentary aquifer and from 2.16×10^{-5} to 4.9×10^{-5} in the hard rock aquifer at a lower level. Decadal fluctuation (1998–2007),

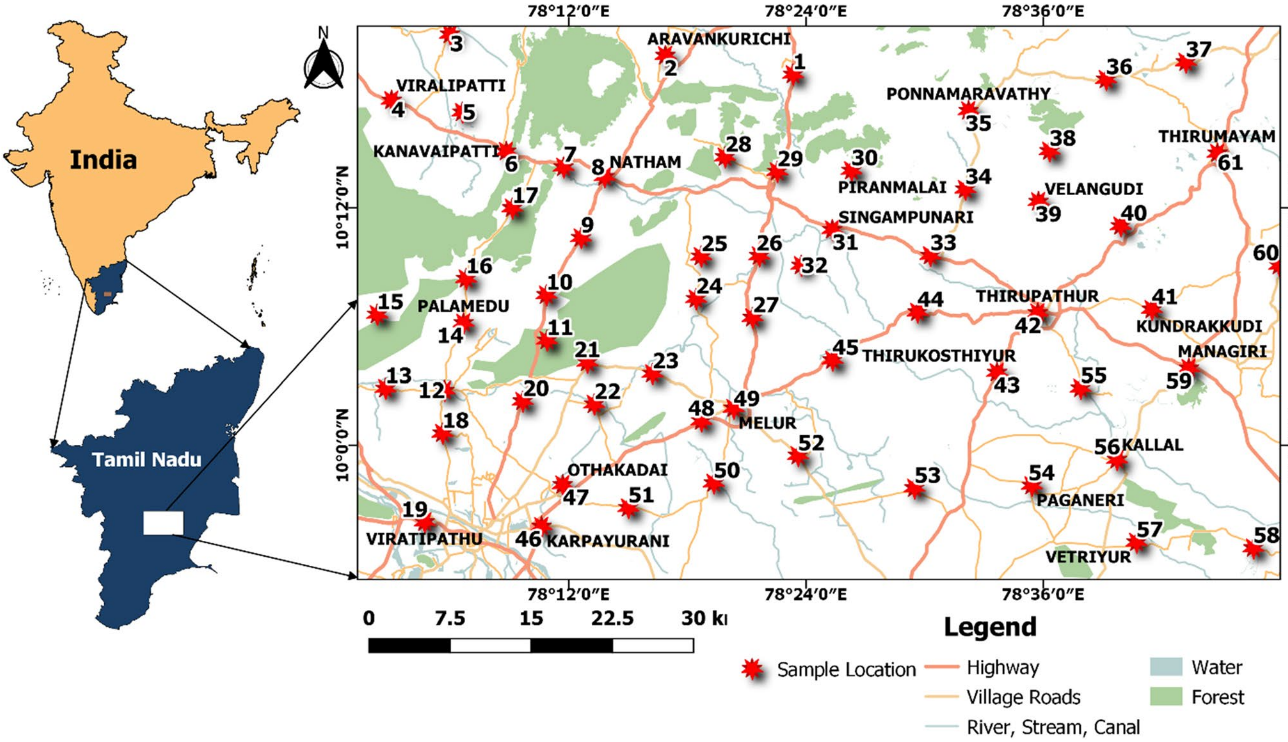

Fig. 1 Location details and sampling points of Central Tamilnadu, India

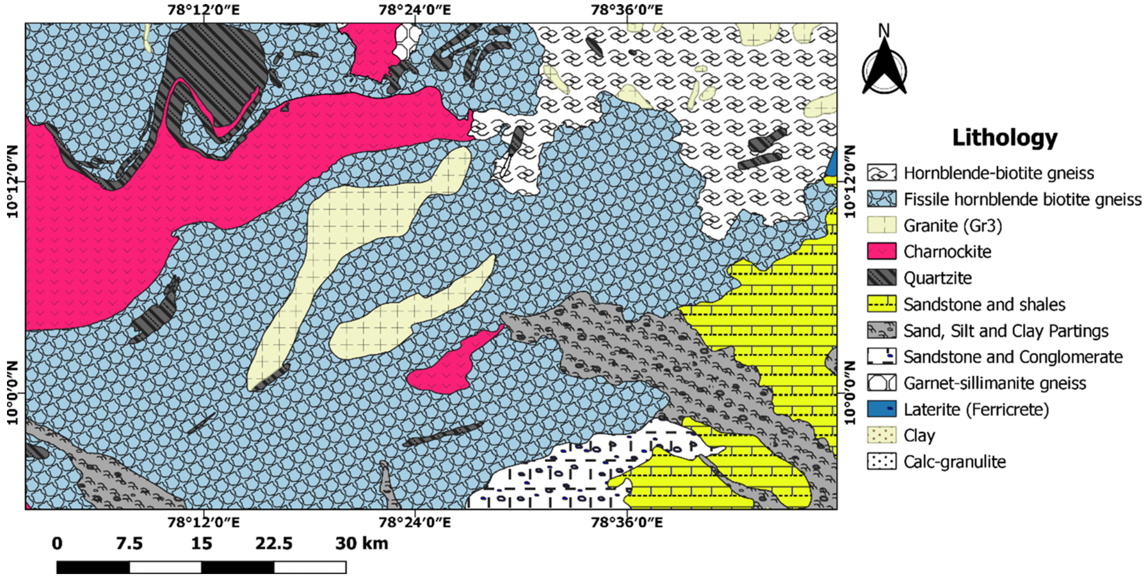

Fig. 2 Lithological distribution in central Tamil Nadu, India

inferred that water table fluctuates from 0.004 to 1.523 m/year. (CGWB 2007). Recharge in the study area is mainly due to the normal rainfall, whereas surface water sources and rate is enhanced by the surface water irrigation. The effect of the rainfall on water levels in nearby region was studied

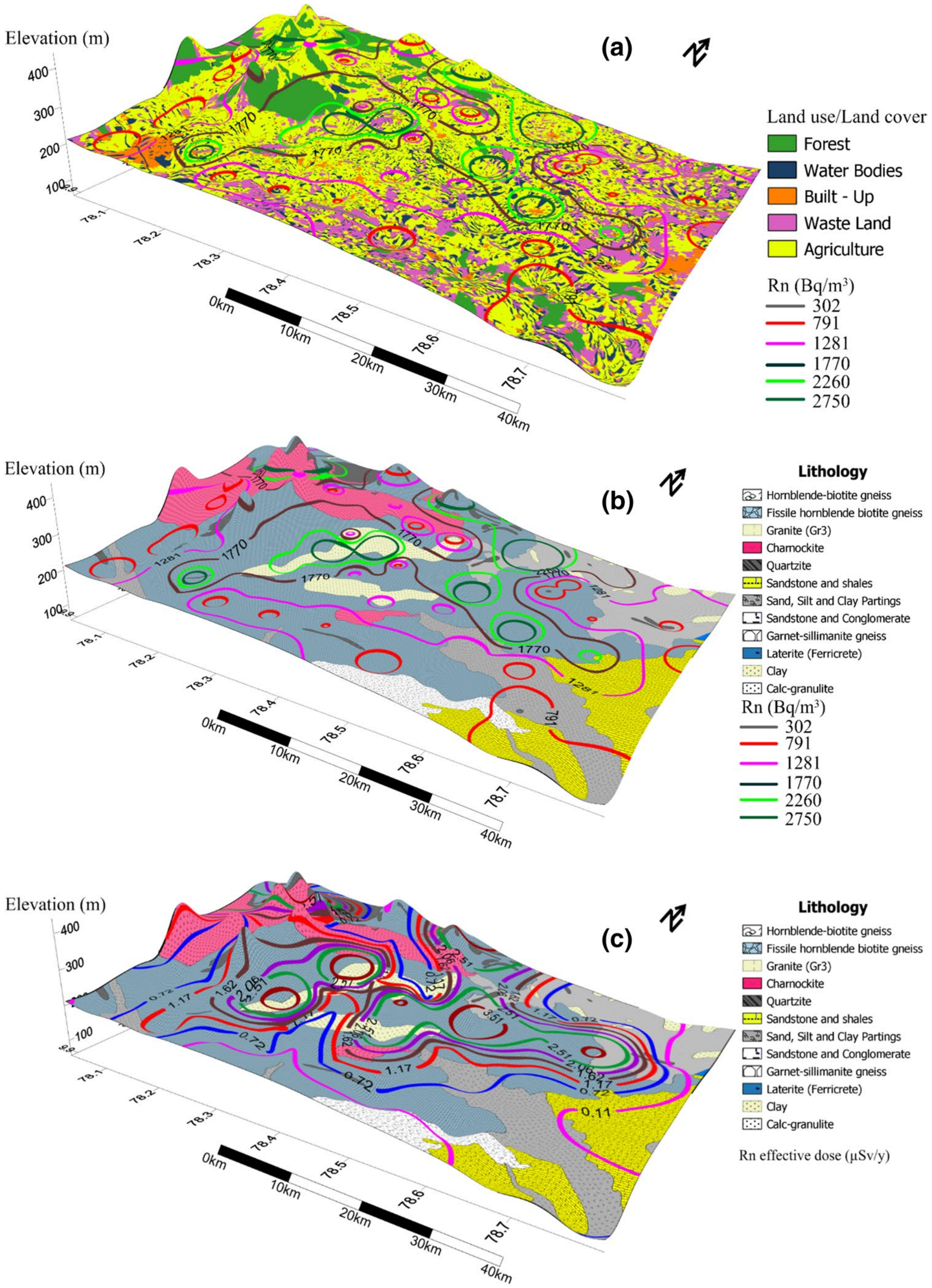

Fig. 3 Spatial distribution of radon over (**a**) land use/land-cover, (**b**) lithology, and (**c**) total annual effective dose of Radon over lithology

by Thivya et al. (2018), Devaraj et al. (2020). Discharge is mainly due to bore well extraction; the groundwater serves as the main source of drinking, domestic, and irrigation purposes in the study region.

Sample Collection and Analysis

58 groundwater samples were collected during postmonsoon period (January 2015) (Table 1) from bore wells, considering spatial coverage and lithology. In situ levels of Rn were measured using portable Smart Radon Monitor (SRM) from Bhabha Atomic Research Centre, India, and the measurement is based on the microprocessor. SRM uses a continuous monitoring technique which provides Rn series of time concentrations in water samples where simultaneous sampling and counting are performed. SRM works based on a software which takes account of the progeny growth in each counting period, without making balance assumptions. The benefit of this technology is that relative moisture does not affect its efficiency, and therefore no silica gel or any other dehumidifying agents are needed (Fig. 4). SRM has been successfully and extensively used for radon and thoron

Table 1 Rn concentration in groundwaters of central Tamil Nadu, India

Sampling number	Location	Rn (Bq/m^3)
1	Puludipatti	3827
2	Aravankurichi	325
3	Kambarpatti	1220
4	Viralipatti	2136
5	Kurumbavetti	308
6	Kanavaipatti	7072
7	Oluppakkudi	3720
8	Natham	605
9	Udukkattai	1142
10	Parali	1476
11	Kadavur-kanvai	1833
12	Alanganallur	258
13	Kalivelipatti	919
14	Palamedu	540
15	Mettupatti	1441
16	Chatravellarapatti	447
17	Chinnamalayur	3711
18	Kumaram	549
19	Viratipathu	302
20	Chattaratondanpatti	1009
21	Thukkalampatti	1981
22	Chettiarpatti	2747
23	Allampatti	4863
24	Periyakarpurapatti	1134
25	Karungalakkudi	6388
26	Manapatti	1078
27	Thanchiyam	1199
28	Kottampatti	460
29	Piranmalai	375
30	Singampunari	2143
31	Nadunattarmangalam	653
32	Marudipatti	4063
33	Ponnamaravathy	5687
34	Kulipirai	2021
35	Nachanthupatti	1117
36	Sevvoor	418
37	Velangudi	292
38	Keezhseralpatti	767
39	Kundrakkudi	2434
40	Thirupathur	4080
41	Thirukosthiyur	828
42	Satrusamharakottai	986
43	Vachampatti (keela valavu)	1067
44	Karpayurani	1317
45	Othakadai	3577
46	Chinnasooragundu	1508
47	Melur	1368
48	Near thiruvadur	593
49	Panaikulam	402
50	Ottapatty	767
51	Keelapungudi	345
52	Paganeri	1054
53	Kundramanikam	469
54	Kallal	295
55	Vetriyur	446
56	Periyakarai	327
57	Pallathur	335
58	Thirumayam	694
	Minimum	258
	Maximum	7072
	Average	1605

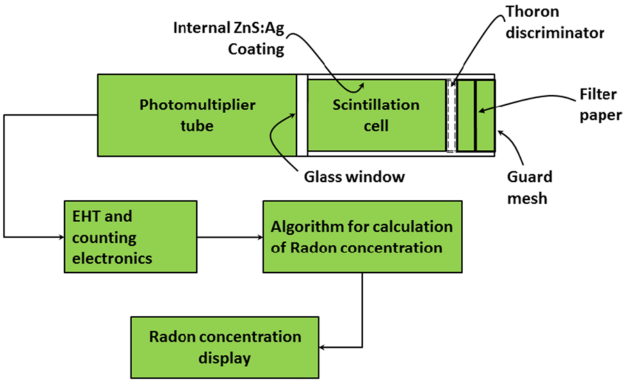

Fig. 4 Portable Smart Radon Monitor (SRM)

detection in different parts of India (Chauhan et al. 2014; Kumar et al. 2016). In Bq/m^3 (disintegration per second per m^3), Rn processes are expressed with 2 sigma uncertainties. SRM displays radon readings after every 15 min of measurement run (after started). The in situ radon readers will take the average readings of 3 or 4 cycles. Using this reading, the level of radon can be determined by correcting the time between the sample collection and analysis using the equation.

$$C = C_0 e^{-\lambda t} \tag{1}$$

where C is the measured level, C_0 is initial concentration (calculated) after the decay correction, t is the time elapsed since collection (days), and $\lambda = (0.693)/(t_{1/2}) = 0.181$, $t_{1/2} = 3.83$ days. The instrument tests the behaviour of radon using the alpha scintillation process. Detection limit of radon is 8 Bq/m^3 at 1 sigma confidence for 1 h counting. The overall calibration accuracy of detector is approximately ± 5%.

Uranium is analysed by laser fluorimeter (Model LF-2a, Quantalase, India). For 15 days, all laboratory glassware used for sample processing was soaked in 10% nitric acid and then thoroughly rinsed with distilled and double distilled water before use. For each batch of sample preparation, the reagent blank was taken and concentrations found in the reagent blank were subtracted from the same batch of samples. The technical features of laser uranium analyser detection limit: 0.2 ppb of uranium; range: 0.5–1000 ppb; excitation source: sealed-off nitrogen laser; wavelength: 337 nm; pulse energy: 20 μjoule; pulse duration: 7 nano second; frequency: 10 Hz and sample size: 3–5 ml.

Data Interpretation

The radiological effects of dissolved radon intake are described as the effective dose of radiation received from the population at the time of daily water intake. A relationship is used to assess an appropriate annual dose for a single person by the ingestion of radon from drinking water (Wu et al. 2014; Krishan et al. 2015).

$$D_W = 1/4\, C_W C_{RW} D_{cW} \tag{2}$$

where D_W is the annual effective dose (mSv/y) due to ingestion of radionuclides from the consumption of water, C_W is the concentration of Rn in the ingested drinking water (Bq L^{-1}), C_{RW} is the annual intake of drinking water (L y^{-1}), and D_{CW} is the ingested dose conversion factor for ^{222}Rn (Sv Bq^{-1}).

The dose conversion metric suggested by the Scientific Committee of the United Nations on the effects of atomic radiation (UNSCEAR 1993) was used to calculate effective dose. Annual effective dose due to Rn consumption was estimated from drinking water, since an adult (age > 18 years) takes an average of 730 L of water per year (WHO 2011). Annual effective doses (mSv/y) and effective doses per liter (mSv/y) were measured following the ingestion of Rn, dissolved in drinking water. Inverse distance weighted (IDW) technique was used for spatial analysis tools in Arc GIS 10.2.

Results and Discussion

Table 1 shows the Rn concentrations in groundwater collected from the 58 sampling locations. Overall, Rn levels ranged from 258 to 7072 Bq/m^3, with an average of 1605 Bq/m^3.

Spatial Distribution of Radon

Spatial distribution of Rn in the study area (Fig. 5) shows that higher-level of Rn were observed in the central and northern portion of the study area. It is further noted that Granitic intrusions are located in the central part. There are few outcrops of Charnockites, Quartzite, Hornblende biotite gneiss (HBG), and mainly fissile hornblende biotite gneiss (FHBG) in the western and northern region of the study area contributing to higher Rn levels compared with the sedimentary formations in the southeastern region (Fig. 3b; Table 2). Lithology, porosity, degree of fracture, flood flow rate, and topography play a major role in the distribution of Rn levels in groundwater (Appleton and Miles 2010; Künze et al. 2013; Pinti et al. 2014; Skeppstrom and Olofsson 2006; Zunic et al. 2014). Groundwater in hardrock aquifer usually contains high Rn because of increased radon migration (Yang et al. 2014; Atkins et al. 2016). Figure 3a depicts the spatial distribution of land use/land cover and Rn levels in the study area revealing their spatial relations between these two parameters.

Higher Rn values were observed around the agricultural lands and the forest areas. Groundwater from wells in

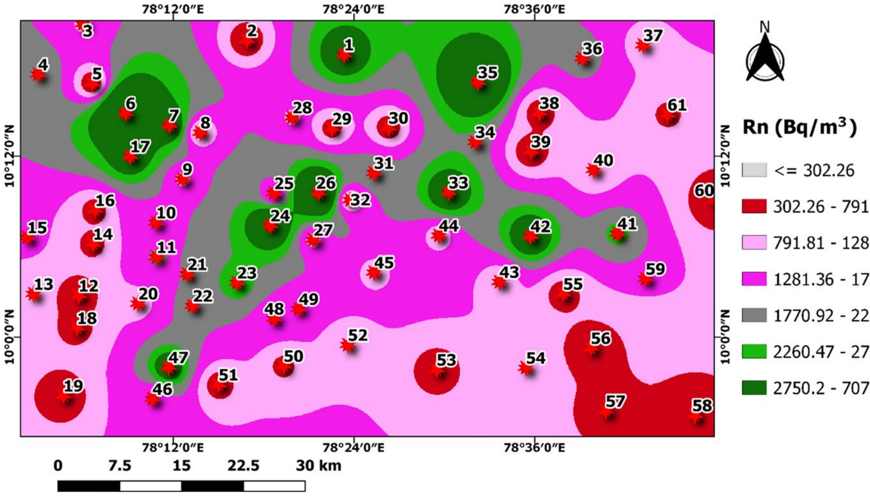

Fig. 5 Spatial distribution of Radon concentration in the study area

Table 2 Variation of Rn concentration in different lithological units

Lithology	Sampling points	Percentage of samples
Clay	Nil	–
Garnet-sillimanite gneiss	Nil	–
Quartzite	1, 61	3.28
Hornblende-biotite gneiss	35, 36, 37	4.92
Laterite (Ferricrete)	Nil	–
Sand, Silt and Clay Partings	19, 43, 55, 56, 58	8.20
Calc-granulite	Nil	–
Charnockite	8, 9, 14, 15, 16, 17, 28	11.48
Fissile hornblende biotite gneiss	2, 3, 4, 5, 6, 7, 10, 11, 12, 13, 18, 20, 21, 22, 27, 29, 30, 31, 32, 33, 34, 38, 39, 40, 41, 42, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54	60.66
Sandstone and Conglomerate	Nil	–
Granite (Gr3)	23, 24, 25, 26	6.56
Sandstone and shales	60, 59, 57	4.92

agricultural settings shows higher levels of U than groundwater from wells in urban settings, possibly due to higher pH influencing the redox levels in agricultural wells (Ayotte et al. 2011).

However, the relationships between Rn level and geological parameters are much more complex than expected due to its short half-life and volatile character (Cho et al. 2015). Sundal et al. (2004) reported that apart from the type of bedrock lithology and structures, Rn levels are also influenced by emanation coefficients, moisture content, permeability, and Rn emission rates.

Relationship Between U and Rn

The scatter plot (Fig. 6) of Rn and U shows that there was a progressive increase in Rn levels with U concentrations, especially in samples collected from granitic terrain. A similar trend was observed in FHBG and Charnockite, indicating the lithological influence in spatial distribution of U and Rn in groundwater. However, the sedimentary formations in the study area do not show any definite trend. The relationship between U and Rn is a good indicator that the groundwater contains a direct geogenic radionuclide source.

U in groundwater of the study area is due to rock water interaction, initiated by the dissolution of U in the host rock and Rn also dissolves into the groundwater during

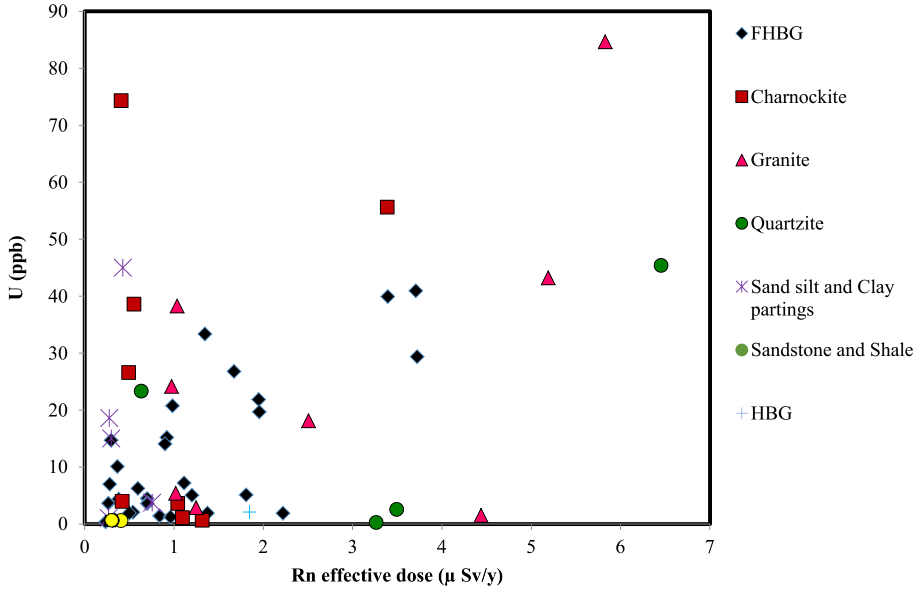

Fig. 6 Correlation between annual effective dose rate of radon (222Rn) in groundwater samples and total U content of the background lithology

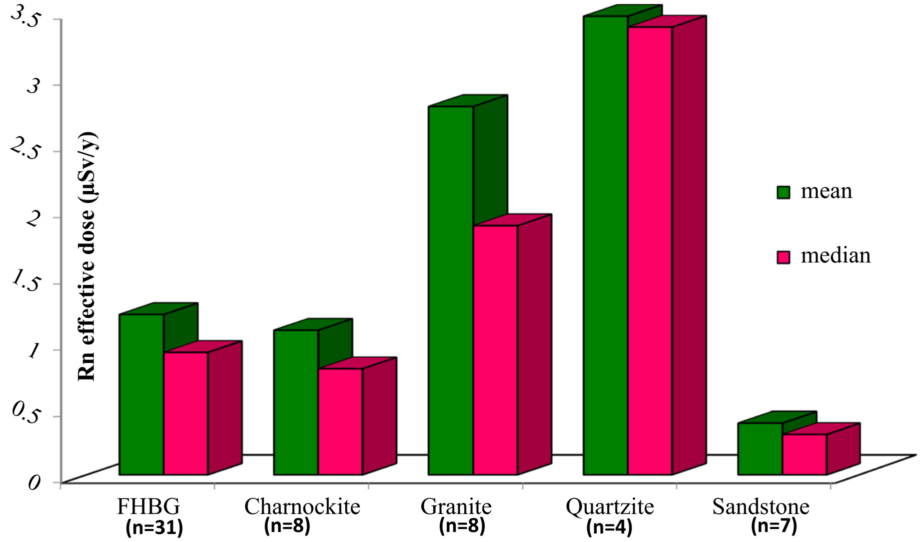

Fig. 7 Distribution of mean and median radon annual effective dose in different lithologies

radioactive decay of U (Arabi et al. 2013). It also has been recorded that U from bedrock is the primary source of Rn (Appleton and Miles 2010; Akerblom and Lindgren 1997). Hence, it is assumed that U is the principal source of Rn in the groundwater of this region (Skeppstrom and Olofsson 2006). Therefore, Rn develops from the lithological U source, concentrated in the fracture zone within Granitic and Gneissic rocks (Choubey et al. 2003). Lithology regulates the distribution of Rn in groundwater (Langmuir 1997), and it is observed that the granite rocks have the highest U and Rn values in the study area. U is commonly found as uraninite in granite, which is easily dissolves to release Rn into groundwater (Kraemer and Genereux 1998).

U is leached from the rock and precipitated on the surfaces of the cracks in the rock along with its decay products, such as radium. Rn is then emanated directly into the groundwater along the fractures from the radium-enriched coatings (Akerblom and Lindgren 1997). The release and movement of radon in the groundwater are regulated by the content of U, grain size, host rock permeability, and the type and extent of fracturing in the host rock (Choubey and Ramola 1997). Granite rocks commonly contain, higher U than sedimentary rocks, such as sandstone (Faure 1986). Adithya et al. (2016) reported higher U concentrations in groundwater samples of hard rock terrain than the sedimentary formations. The period of groundwater residence in Granite, Charnockite, and Gneissic rocks facilitates the greater release of U into groundwater (Adithya et al. 2016).

It was noted by earlier researchers that U need not be enhanced by a single factor as observed by lithological influence (Thivya et al. 2017). The highest dissolution of U occurs in near-neutral to acidic groundwater pH conditions. In addition, to other responsible sources, redox potential also is a major governing factor that contributes to higher U in groundwater (Harley and Robbins 1994). This clearly shows that the concentration of Rn depends primarily on the lithology, structural attributes, the nature of U minerals in rocks, and the conditions of redox.

Effective Dose Assessment

The global average dose of radon inhalation and its decay products from all sources is approximately 1 mSv/y (Crawford-Brown 1989), which is significantly less than half the overall exposure of 2.4 mSv/y to natural radiation (NRC 1999). Similarly, the total annual effective dose of radon, and the annual effective dose in the groundwater sampled, varied with radon levels. In the present research, the annual effective dose of radon ranged from 0.24 to 6.45 mSv/y, with an average value of 1.46 mSv/y. The overall effective annual dose (mSv/y), resulting from radon in groundwater of the study area was substantially lower than the acceptable level of 1 mSv/y for the public (EPA 1998).

The annual effective radon dose in groundwater is dependent on the total U content of the host rock formations. The samples from granitic terrain show that there is a progressive increase in Rn dose with higher U. Similar trend also was observed in FHBG and Charnockite rocks. There was no definite trend between U and effective dose of Rn in the sedimentary formations of the study area. It is thus apparent that there is a clear correlation between the overall annual effective dose of radon in groundwater and the host rock U content (Skeppstrom and Olofsson 2006), suggesting the influence of background lithology on the radiological properties of the groundwater.

The spatial variation of the total annual effective dose of Rn in the groundwater samples (Fig. 3c) shows that the northern and central portion of the area exhibits higher levels. This sector was represented by Granite, Charnockites, FHBG, and Hornblende-biotite gneiss rocks. Observed elevated Rn levels are explained by enhanced Rn migration through fractures and weaker planes of the host rocks (Damkjær et al. 1997; Wood et al. 2004). In the case of the southern and eastern parts of the area, low Rn levels in groundwater are observed, as is common in sedimentary rocks (Cothern and Smith Jr. 1987).

The mean and median values of Rn effective dose is shown in Fig. 7. The Rn effective dose with respect to the sampling points exhibit the following order; Quartzite (*n*−4) > Granite (*n*−8) > Charnockite (*n*−8) > Fissile Hornblende Biotite Gneiss (*n*−31) > Sand, Silt, and Clay partings (*n*−7). This also clearly states that Rn levels in groundwater are dependent on the lithology of the study area. Rn activity is typically low in sedimentary rock units and higher in Granite, FHBG, and Charnockite terrains (Cothern and Smith Jr. 1987).

There is more difficulty with respect to the Rn levels in a fractured aquifer since, given that the flow is greater in the fracture, due to the heterogeneous nature of a fractured granitic aquifer, the Rn level is variable (Le Druillennec et al. 2010). In addition, groundwater in India was found to have no clear association between Rn and U and no normal pattern in the difference between Rn and the source of water (Singh et al. 2009).

Conclusions

Based on the results of this study, the Rn concentration was observed to be below the maximum contaminant level (MCL) in all groundwater samples. Overall, the Rn levels in sedimentary rock formations displayed lower value. Higher concentration of Rn was mostly observed in the central and NW region of the study area, due to the release of Rn from granite, gneissic rocks, and quartzite formations. Rn levels in groundwater is due to high mobility of Rn facilitated by suitable geological conditions, apart from other factors, including agriculture. It is inferred that granite rocks were the main source of Rn in the groundwater, associated with U dissolution. This observation indicates that Rn was co-transported with U into U-enriched zones. The effective annual dose values also were significantly lower than 1 mSv/y and were well below the UNSCEAR and WHO guidance values. Rn in groundwater of the central region of Tamilnadu has exceptionally low levels and do not pose a significant health risk. However, the Rn level may be increased in the groundwater in near future due to the intensive agricultural practice in the study area. Hence, the policy makers need to have a continuous monitoring on Rn levels in the groundwater, in order to take necessary steps to safeguard this water resource from pollution. The data presented are limited to 58 groundwater samples representing a particular season, which must be treated as a preliminary observation. Therefore, to improve the understanding and reduce potential risks, more comprehensive research should be undertaken by detailed monitoring during various seasons for possible contamination of Rn.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

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