



Monitoring geothermal springs and groundwater of Pir Panjal, Jammu and Kashmir, for radon contamination

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Abstract

Hot water springs and bore wells/hand-pumps were investigated to quantify radon and uranium levels in Rajouri area of the Pir Panjal. Scintillation-based radon monitor was employed for radon-222 detection while as LED Fluorimetric technique was used to detect uranium-238 concentration. The radon-222 levels, found in the study area, are much higher than the limits prescribed by regulatory agencies like United States Environmental Protection Agency (USEPA). Some of the samples exceeded the allowed limits of 100 Bq L⁻¹ set by World Health Organisation while none of the samples lied within the prescribed level of 11 Bq L⁻¹ prescribe by USEPA.

Keywords Groundwater radioactivity · ²²²Rn · ²³⁸U · Pir-Panjal · Geo-thermal springs · Effective dose

Introduction

Pir Panjal, rich in faults and fractures, is known for geothermal springs which are known to have high radon concentrations [1, 2]. Surface water and groundwater are two major sources of drinking water on the planet. In fact, groundwater contributes to almost half of all drinking water, globally [3]. During its course, groundwater comes in contact with various radium rich rocks and thus, radon gets dissolved in it [4, 5]. It is, therefore, imperative to measure radioactive contamination present in groundwater. Higher concentration of radon-222 in groundwater is an indicator of higher levels of indoor radon as soil and building materials are the largest sources of indoor radon. Airborne radon and its decay products contribute to an average inhaled dose of 1.26 mSv per year which is almost half of the natural background radiation dose received by the general public [6] and therefore poses an immense health risk [7–9]. Although, ²²²Rn delivers dose

due to both ingestion as well as inhalation, nevertheless, the latter is the dominant factor to the dose received by humans.

The occurrence of natural radionuclides depends on several factors like lithology and presence or absence of faults and fractures [10–13] and therefore the concentration of these radionuclides have a huge disparity throughout the surface of the earth. Uranium-238 is the most abundant isotope (99.28%) and is found in trace amounts almost everywhere in earth's crust. ²³⁸U has a half-life of 4.5 billion years and therefore is not a highly radioactive element. In fact, at high concentrations, its chemical toxicity by far exceeds its radiological toxicity [14, 15]. The ²³⁸U decay series ends with the stable ²⁰⁶Pb isotope. In the decay series of ²³⁸U, all elements but radon-222 are solids.

This study is focussed at the radiological threat posed by ²²²Rn to the local population [7] as it is a major contributor (> 50%) of the background dose received by humans due to ionizing radiation [6]. Out of the various isotopes of radon, ²²²Rn (half-life of 3.8 days) is most significant followed by ²²⁰Rn (half-life of 56 seconds). In the present study, ²²⁰Rn has been neglected due to its relatively short half-life. Radon is a known carcinogen and holds the reputation of being the second most important cause of lung-cancer, next to smoking [9, 16]. It is radioactive, inert, tasteless and colourless. Thus, even if it may be present in the indoor environment, our sensory organs cannot sense it which makes it obligatory to detect, quantify and prevent exposure due to it. Interestingly, it is the radon progeny, which are more dangerous than

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the gas itself. Being a noble gas with a short half-life, radon neither reacts with the surrounding environment nor does it accumulate for long. However, its progeny attach themselves to aerosols suspended in the air and can, therefore, enter into the lungs where they can either be retained by the lungs or enter the bloodstream, thus contributing significantly to the internal radiation dose [17–19].

Several inspiring research works have been published to assess the water quality of the study area [20–22]. However, to our knowledge, the present study is a first to assess radiation dose rates due to radon-222 in drinking water. Additionally, the importance of the study comes from the fact that very little or no work has been carried out to reckon the radioactive contamination in the study region [23–25] and therefore this study fills the gap of the data available from the region.

Materials and methods

Study Area

Rajouri district, falling in the Pir Panjal mountain range, is primarily a mountainous zone-Dera Gali, Pir Panjal and Rupri pass being some of the noticeable peaks in the area [26]. Figure 1 shows a map of the study area where it can be seen that the area lies between the Main Boundary Thrust (MBT) and the Riasi Thrust. The altitude of the region varies from 460 to 3900 m asl. As per Census 2011,

Rajouri has a total population of 0.64 million with an area of 2,769 km². The main lithological formations include the Siwalik formation, Murree, Panjal Trap, Agglomerated slate, Baila and Gamir formation. Gamir and Baila formations are predominantly composed of black carbonaceous and calcareous shale, shale-slate combination with lenticular bands of limestone. Panjal Trap, which succeeds the Agglomerated slate, varies from greyish green to dark green in colour. Limestone and coal are the main minerals of the economic importance in the district occurring mainly in the Subathu formation [27]. The main source of drinking water to the local populace comes from surface and groundwater sources like springs, wells and boreholes which collectively comprise of about ~58% of the drinking water source while as tap-water is supplied to only about 32% of the households. Due to the socioeconomic constraints, only about one-third (32.98%) of the population live in permanent dwellings while as the majority (64.44%) live in semi-permanent houses with improper ventilation conditions [28].

Sample collection and analysis

Samples were collected in 60 mL glass bottles provided with the detector and the cap of the bottles was tightened underwater to ensure zero radon loss. It has been observed that there is least radon loss in glass bottles as compared to other materials [29]. Also, in-situ analysis of all the water samples was carried out to ensure better results.

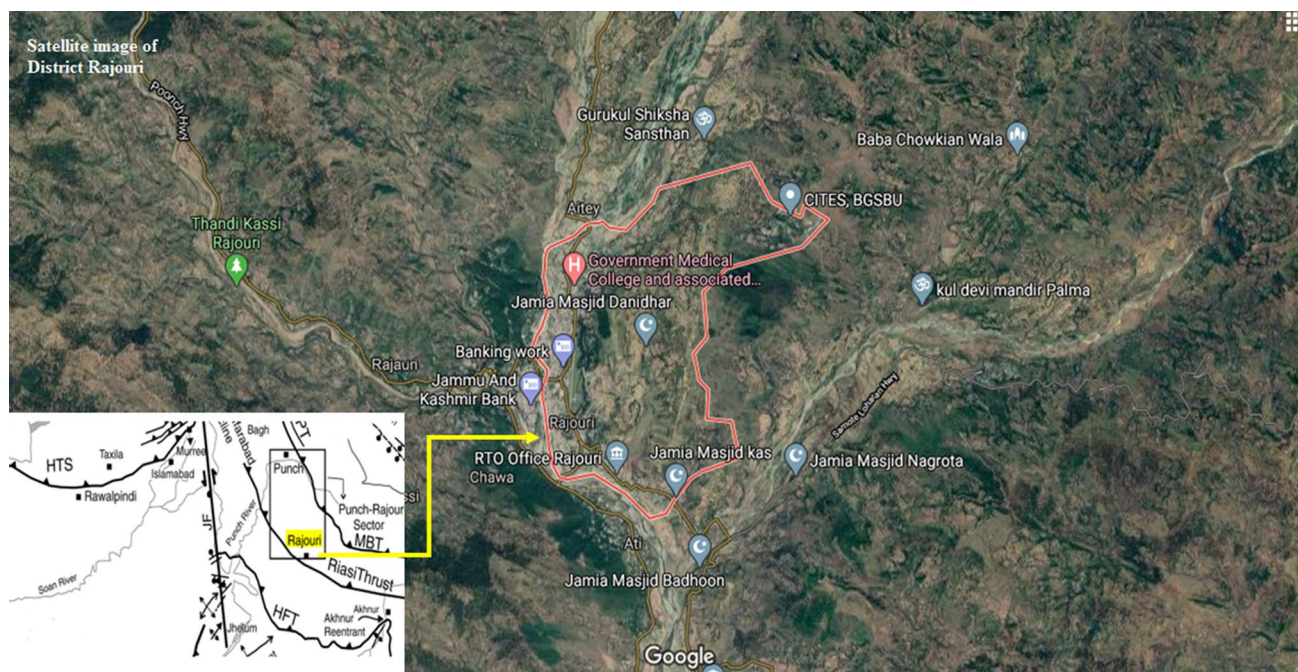


Fig. 1 A satellite image of the study area

Measurement of radon activity concentration

The survey was carried between May and July 2019 at Rajouri, Pir Panjal, using a scintillation based radon monitor (RnDuo, AQTEK Pvt. Ltd. India) developed by Bhabha Atomic Research Centre, Mumbai, India [30]. The RnDuo is a portable continuous activity monitor for measurement of radon (^{222}Rn), thoron (^{220}Rn) and gross alpha from the sampled gas. This device is based on the principle of detection of alpha particles emitted from sampled radon and its decay products formed inside the detector by scintillation with ZnS: Ag. The experimental procedure consists of a water bubbler provided along with the scintillation detector. Before taking the measurement of each sample, the detector volume was flushed for 5 min in an open-loop using the intrinsic pump of the detector so as to remove any residual gas present in the scintillation cell. After the water bubbler was connected to the detector, the detector was operated in a 15 min cycle for 1-h such that for each sample, four measurements were recorded. The average value was taken as the true radon concentration of the sample. The procedure was repeated for each sample. The lower detection limit of radon for the monitor is 8 Bq m^{-3} at 1 sigma and 1 h counting cycle while as the upper detection limit is 10 MBq m^{-3} . To ensure precision in the measurements, each year the detector is taken to a facility at Bhabha Atomic Research Centre (BARC), Mumbai, India, for calibration purposes. Furthermore, the detector has no effect of humidity which gives it an advantage over other commercially available scintillation detectors. Scintillation detection technique was preferred

over other methods because of its greater efficiency than other passive methods [31]. A schematic of the instrument is shown in Fig. 2.

The dissolved radon-222 concentration in groundwater was calculated from the concentration measured in air (pumped in the detector-volume of the radon monitor) by using the equation:

$$C_w = C_a \times (K + V_a/V_w) \quad (1)$$

where C_w is the concentration of ^{222}Rn in water, C_a is the concentration of ^{222}Rn in air, K is a dimensionless quantity called as partition co-efficient of radon in liquid with respect to air ($=0.25$ for water), V_a and V_w are the volumes of air and water respectively [32, 33].

Uranium measurement employing LED fluorimeter

For the estimation of uranium-238 concentration in groundwater, LED fluorimeter (manufactured by Quantalase Enterprises Pvt. Ltd., Indore, India), was used. The functioning of this instrument is based on the fluorescence of uranyl ions present in the aqueous solution and this technique is considered as the most reliable, quickest and efficient for the uranium estimation up to ppb or sub-ppb level in the aquatic environment. The detection limit of this instrument is from 0.2 to $1000 \mu\text{g L}^{-1}$.

The instrument consists of UV LEDs as an excitation source having a wavelength of 400 nm , compartment for holding the sample and the photomultiplier tube (PMT).

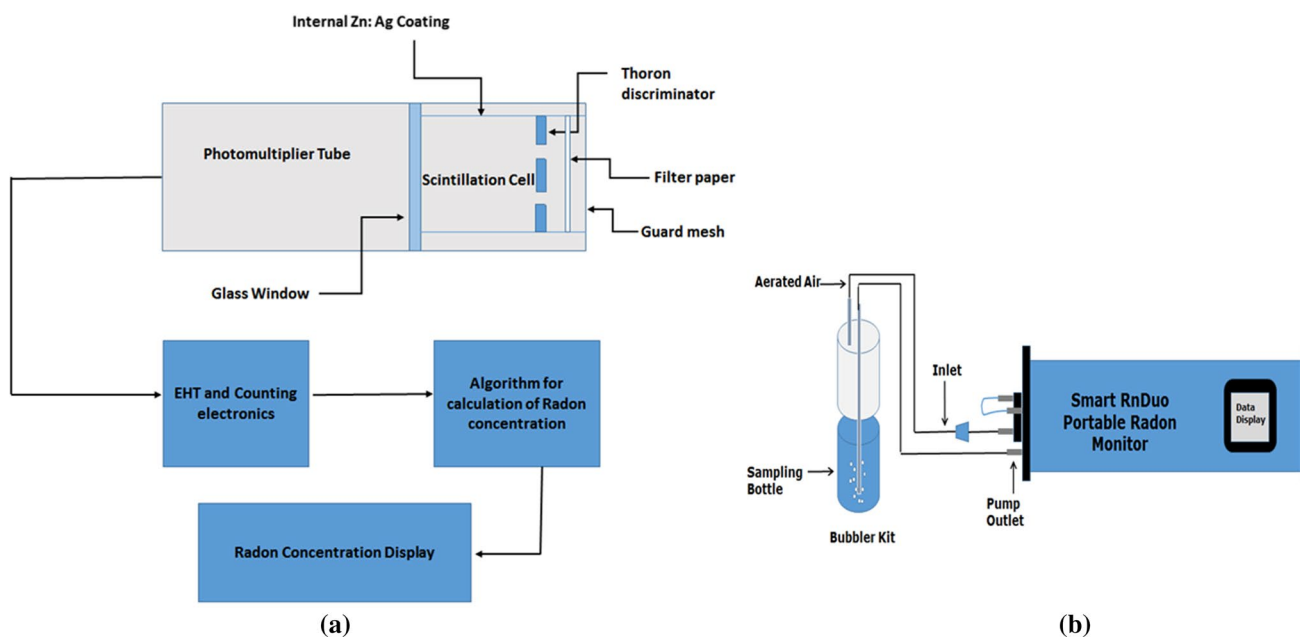


Fig. 2 A schematic of the scintillation detector

To obstruct the fluorescence from organic matter to fall on PMT, suitable filters are placed in the middle of excitation source and PMT. Additionally, proper time-gating of PMT is done to avoid the fluorescence from organic matter to fall on PMT which has short lifetime of ~ 100 ns whereas fluorescence from uranyl ions is ~ 200 μ s.

5% of sodium pyrophosphate was prepared in double-distilled water which acts as fluorescence-enhancement reagent. The pH of this solution was adjusted to 7 by adding ortho-phosphoric acid dropwise and then it was added to groundwater samples in the ratio of 1:10 to change all uranium species into single uranyl phosphate complex so as to obtain same fluorescence yield. Before uranium-238 estimation in groundwater samples, the instrument was calibrated with 10 μ g L⁻¹ uranium-standard solution which was prepared by diluting 10⁵ μ g L⁻¹ uranium-standard solution supplied by Quantalase Enterprises Pvt. Ltd.

Estimation of dose due to radon concentration in water

Groundwater, contaminated with ²²²Rn, delivers inhalation dose to lungs and ingestion dose to various organs most notably the stomach. The inhalation and ingestion doses were calculated by using the equations given by UNSCEAR [34–36] as:

$$D_{inh} = {}^{222}\text{Rn} \times \varphi \times F_R \times DCF_R \times T_{occ} \quad (2)$$

$$D_{ing} = {}^{222}\text{Rn} \times AI_W \times DCF_{ing} \quad (3)$$

where D_{inh} and D_{ing} are the inhalation and ingestion doses due to radon-222, respectively. ²²²Rn is the concentration of radon in water, φ is the air to water concentration ratio (10⁻⁴), and F_R (=0.4) is the equilibrium factor between ²²²Rn and its progenies [37, 38]. DCF_R is the dose conversion factor for ²²²Rn inhalation (9 nSv (Bq h m⁻³)⁻¹) while as T_{occ} is the mean indoor occupancy time (~ 7000 h per year). DCF_{ing} is the dose conversion factor for ingestion (3.5 nSv Bq⁻¹) while as AI_W is the age-wise daily water intake.

Results and discussion

An analysis of the data revealed that the groundwater is contaminated with ²²²Rn. The most contaminated region was Kalakote which is famous for its coal mines.

As shown in Table 1, all of the samples exceed the safe levels of 11 Bq L⁻¹ set by USEPA while as 40% of the samples exceed the safe levels of 40 Bq L⁻¹ set by UNSCEAR and ~ 27% of the samples exceed the safe limits of 100 Bq L⁻¹ set by the WHO. The descriptive statistics of ²²²Rn and ²³⁸U concentrations are provided in Table 2.

Table 1 Radon concentration and the associated inhalation dose along with the uranium concentration in geo-thermal springs and groundwater of the study area

Sample code	Source	Radon concentration (Bq L ⁻¹)	Inhalation dose (mSv year ⁻¹)	Uranium (μ g L ⁻¹)
R-11-01W	Spring	32.8 ± 1.3	0.08	0.38
R-11-01W	Spring	131.0 ± 2.7	0.33	0.34
R-11-01W	Spring	14.6 ± 0.9	0.04	BDL
R-11-01W	Spring	189.2 ± 3.0	0.48	0.19
K-16-05W	Spring	149.7 ± 3.2	0.38	0.34
K-16-06W	Spring	126.8 ± 3.1	0.32	BDL
K-16-12W	Spring	19.1 ± 1.0	0.05	0.16
K-16-13W	Spring	51.4 ± 1.7	0.13	0.14
K-16-15W	Hand pump	23.5 ± 1.3	0.06	BDL
K-16-16W	Hand pump	44.6 ± 1.5	0.11	0.56
K-16-17W	Hand pump	37.5 ± 1.6	0.09	2.9
K-16-18W	Hand pump	15.6 ± 1.1	0.04	1.4
K-16-19W	Hand pump	24.9 ± 1.2	0.06	3.2
K-16-20W	Hand pump	21.4 ± 1.3	0.05	1.1
K-16-21W	Hand pump	27.1 ± 1.1	0.07	1.5

BDL below detection limit

Table 2 Descriptive statistics of radon and uranium found in water samples collected from the study region

Statistical parameter	Radon (Bq L ⁻¹)	Uranium (μ g L ⁻¹)
Mean	60.61	0.8
Standard error	14.90	0.27
Median	32.80	0.34
Standard deviation	57.72	1.03
Kurtosis	0.19	1.44
Skewness	1.28	1.53
Range	174.60	3.2
Minimum	14.60	BDL
Maximum	189.20	3.2
Count	15.00	15

The concentration of ²²²Rn varied from a minimum of 14 ± 0.90 Bq L⁻¹ to a maximum of 189 ± 3 Bq L⁻¹ with an average value of 60 Bq L⁻¹ while as the concentration of ²³⁸U varied from ‘below detection limit’ to a maximum of 3.2 μ g L⁻¹ with an average value of 0.8 μ g L⁻¹. Figure 3 shows a frequency distribution of the ²²²Rn concentration in the Rajouri region of Pir Panjal. It can be seen that at least 4 samples exceed the allowed limits of 100 Bq L⁻¹ set by WHO while none of the samples lies within the safe limits of 11 Bq L⁻¹ prescribe by USEPA.

The high values of radon-222 concentration is attributed to the geology of the region. Physio graphically, the area is

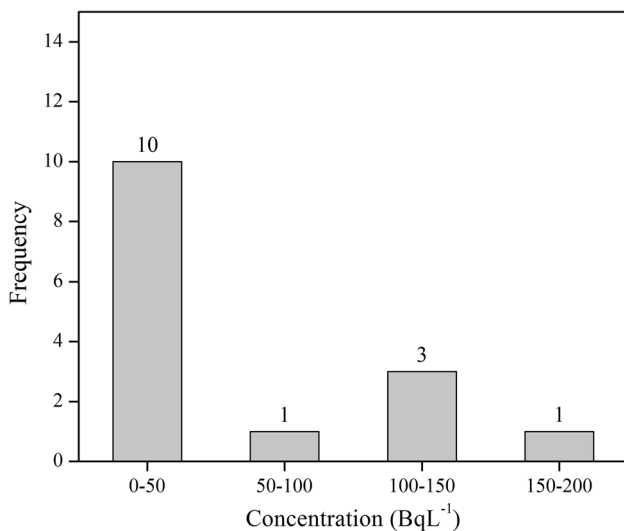


Fig. 3 A frequency distribution of the radon concentration of the samples

situated within the Murree–Siwalik range and to the south of the Pir Panjal range. Structurally, it lies between the Riasi Thrust in the south and the Main Boundary Thrust (MBT) in the north which explains the exceptionally higher values of radon (²²²Rn) in the area [39].

The inhalation dose varied from 0.04 to 0.48 mSv year⁻¹ with an average of 0.15 mSv year⁻¹. The ingestion doses for various age groups have been tabulated in Table 3. The maximum, minimum and average values have been marked bold. It can be observed that the minimum ingestion (0.04 mSv year⁻¹) dose was found to be in infants (0–6 months) and the maximum ingestion dose (2.62 mSv year⁻¹) was found to be received by lactating mothers. The average values (0.15–0.84 mSv year⁻¹), tabulated in Table 3 are less than the recommended level of 1 mSv year⁻¹ for frequent exposure prescribed by the ‘National Council on Radiation Protection and Measurements’ [40]. Table 4 shows the radon-222 concentration in different areas worldwide.

Kolmogorov–Smirnov test and Shapiro–Wilk test both revealed that the radon values are statistically significantly different than a normal distribution and thus not normally distributed.

Figure 4a, b show a Box–Whisker plot of radon-222 and uranium-238, respectively. The upper and lower whiskers represent the minimum and maximum values of concentration respectively while the solid triangle within the box represents the mean value. The lower and the upper lines of the

box denote the 1st and the 3rd quartile respectively while the line within the box denote the median or the 2nd quartile.

Uranium-238 concentration, as analysed in groundwater samples collected from Rajouri area using LED fluorimeter and their concentration at different sampling sites are given in Table 1. The uranium-238 concentration varies from BDL to 3.2 µg L⁻¹, which is quite less and fall below the safe limit of 30 µg L⁻¹ recommended by WHO (2011) [41]. So, the uranium-238 concentration observed in the collected groundwater samples did not show much variation, indicating the even distribution of natural uranium within a safe limit in the study area. The lack of data on radium limits the scope of this study. However this flaw will be rectified in future, larger studies in the area.

Conclusion

- The population of the Pir Panjal area should avoid direct consumption of groundwater. In case of unavailability of tap water, the standard protocol for radon mitigation must be followed. The easiest, economical and most effective method is to simply agitate the water collected from groundwater sources by pouring it from one container to the other container. This would remove most of the radon gas present in the water. The container used to store water should be preferably kept without a lid.
- In the present investigation, radium content in drinking water was not quantified which results in a major flaw in the study. Therefore, radium content must be measured in future assessments, as its quantification is crucial for all radon mitigation methods.
- The administration should set up state programmes for radon mitigation in drinking water. These programmes may include various measures like indoor radon testing, radium quantification in groundwater, public awareness, financial incentive programs or any other assistance required for radon mitigation.
- Uranium-238 concentration in the groundwater samples were found within the permissible value of 30 µg L⁻¹.
- A more detailed study, with larger data points, is needed to better understand the radon distribution in the Pir Panjal region. In future, indoor radon assessment using active and passive techniques should be carried out on a large scale to map the spatial distribution of indoor radon. In addition, radium content in water should be studied.

Table 3 Age-wise ingestion dose due to radon concentration in Rajouri district

Life stage	Ingestion dose for various age-groups (mSv year ⁻¹)											
	Infants (0–6 months)	Infants (7–12 months)	Infants (1–3 year)	Children 4–12 year)	Males (9–13 year)	Males (14–18 year)	Male (adults) (9–13 year)	Females (9–13 year)	Females (14–18 year)	Females (adults)	Pregnancy	Lactation
	0.08	0.10	0.16	0.20	0.29	0.40	0.44	0.25	0.28	0.32	0.36	0.45
	0.33	0.38	0.62	0.81	1.15	1.58	1.77	1.00	1.10	1.29	1.43	1.82
	0.04	0.04	0.07	0.09	0.13	0.18	0.20	0.11	0.12	0.14	0.16	0.20
	0.48	0.55	0.90	1.17	1.66	2.28	2.56	1.45	1.59	1.86	2.07	2.62
	0.38	0.44	0.71	0.93	1.31	1.80	2.02	1.15	1.26	1.48	1.64	2.08
	0.32	0.37	0.60	0.79	1.11	1.53	1.71	0.97	1.06	1.25	1.39	1.76
	0.05	0.06	0.09	0.12	0.17	0.23	0.26	0.15	0.16	0.19	0.21	0.26
	0.13	0.15	0.24	0.32	0.45	0.62	0.69	0.39	0.43	0.51	0.56	0.71
	0.06	0.07	0.11	0.15	0.21	0.28	0.32	0.18	0.20	0.23	0.26	0.33
	0.11	0.13	0.21	0.28	0.39	0.54	0.60	0.34	0.37	0.44	0.49	0.62
	0.10	0.11	0.18	0.23	0.33	0.45	0.51	0.29	0.31	0.37	0.41	0.52
	0.04	0.05	0.07	0.10	0.14	0.19	0.21	0.12	0.13	0.15	0.17	0.22
	0.06	0.07	0.12	0.15	0.22	0.30	0.34	0.19	0.21	0.25	0.27	0.35
	0.05	0.06	0.10	0.13	0.19	0.26	0.29	0.16	0.18	0.21	0.23	0.30
	0.07	0.08	0.13	0.17	0.24	0.33	0.37	0.21	0.23	0.27	0.30	0.38
Minimum	0.04	0.04	0.07	0.09	0.13	0.18	0.20	0.11	0.12	0.14	0.16	0.20
Maximum	0.48	0.55	0.90	1.17	1.66	2.28	2.56	1.45	1.59	1.86	2.07	2.62
Average	0.15	0.18	0.29	0.38	0.53	0.73	0.82	0.46	0.51	0.60	0.66	0.84
Median	0.08	0.10	0.16	0.20	0.29	0.40	0.44	0.25	0.28	0.32	0.36	0.45
Standard deviation	0.15	0.17	0.27	0.36	0.51	0.70	0.78	0.44	0.48	0.57	0.63	0.80

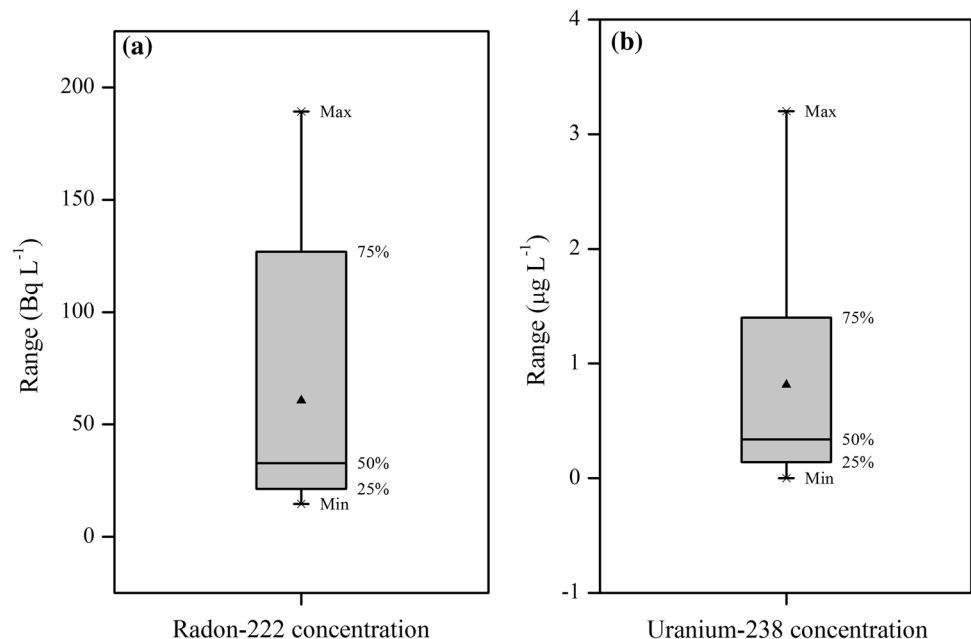
Table 4 Radon concentration (Bq L^{-1}) in drinking water samples reported worldwide

S. no.	Country	Radon concentration (Bq L^{-1})	References
1.	China	0.71–3735 (229.4)	[42]
2.	Brazil	< 1.2–3542	[43]
3.	SW Poland	0.2–1645 (240)	[44]
4.	Finland	27–460	[46]
5.	South Korea	50–300	[47]
6.	Rajouri, Jammu Kashmir	14.6–189.20 (60.6)	Present study
7.	Saudi Arabia	0.01–67.44	[48]
8.	Lebanon	0.46–49.6	[10]
9.	Srinagar, Jammu Kashmir	0.02–38.5	[33]
10.	Khyber Pakhtunkhwa, Pakistan	1.6–18.2	[49]
11.	Australia	(0.14–20.33 (4.90))	[50]

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References

Fig. 4 A Box–Whisker plot of the concentration of radon-222 and uranium-238 in the study area



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Author contributions SN and SS designed the research plan and wrote the manuscript with contributions from BKS, RM, TS and SM. SN carried out all the radioactive surveys. SN and TS analysed the samples. All authors read and approved the final manuscript.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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