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# Estimation of Exhalation Rates of Radon/Thoron in Soil in Hanumangarh District of Northern Rajasthan, India

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Radon and thoron are naturally occurring radioactive gases that are released by geological minerals. If the levels of these gases are high, then their inhalation is closely related to probability of a lung cancer. In present study, radon mass and thoron surface exhalation rates have been evaluated using active radiation detector (Smart RnDuo). It was found that radon mass exhalation rates have lower values while as thoron surface exhalation rates have higher values than the recommended values. The study is valuable as it estimates the radiation contamination of soil, the primary source of contribution towards radon.

**Keywords:** Radon; Thoron; Exhalation rates; Smart RnDuo; Radioactivity

## 1 Introduction

Radon which is a radioactive inert gas originates from rocks and soils and tends to accumulate in closed spaces like houses or underground mines. The primary source of radon in houses is known to be soil-gas infiltration. As a result of the existence of minerals rich inU-238 and Th-232, radon and thoron are typically produced in soil. In a similar manner, the building materials that contain these minerals likewise serve as sources. In the crust of the earth or any porous matrix, radon or thoron is only free to travel if its atoms make their own way into pores or capillaries. There are two ways that radon enters indoor environments through the matrix of soil and building materials. The process by which radon atoms are released into the air-filled pores from solid mineral-grains is known as emanation. Exhalation causes radon gas to migrate from air-filled pores to the atmosphere. The residents inhale radon that is released from the construction materials. Numerous factors including temperature, moisture content, and the activity concentration of radionuclides in rock and soil, affect the pace of radon emanation[1]. Human activity, particularly house construction, has the potential to increase or decrease radon and thoron exposure. The effect of temperature on radon emission is not as significant as the effect of moisture content. This is due to the higher likelihood of a radon atom entering a pore that is completely or partially filled with water being halted in the pore volume and expelled into the atmosphere. The National Academy of Sciences (USA) conducted an extensive review on the Biological Effects of Ionising Radiations (BEIR, VI) and concluded that long-term exposure to radon and its progenies can have detrimental effects on the population including pulmonary disorders especially development of lung carcinoma[2-3]. Thus, quantification and proper mitigation of the radon release from the construction materials is therefore essential.

Based on extensive research into radon and its progenies effects on uranium miners, the International Agency for Research on Cancer (IARC) declared radon to be a human carcinogen in 1988[4]. A few decades later, the World Health Organization (WHO) determined that radon, which accounts for 3–14% of all lung malignancies, is the second-leading cause of lung cancer after smoking. This was based on the very vast studies on radon being conducted in China, Europe, and North America which showed a clear connection between indoor radon exposure and lung cancer[5-12]. Under-lying soil of the dwelling is the principal origin of indoor radon. Despite typically contribution of 1% of the indoor air, soil gas contributes as much as 10% of the indoor air in homes with inadequate foundations or permeable soil (many

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cracks, poor floor-to-wall joints, etc.)[13-15]. Due to emerging energy-saving and energy-efficiency trends in the building industry, the contribution of radon from building materials has recently become more significant. Owing to the modernisation and the need for better thermal insulation, the previously used wooden-framed windows are rapidly being changed with the likes of Unplasticized Polyvinyl Chloride (UPVC) based windows[16]. This, even though greatly help in thermally insulating the building, but significantly slows down air exchange, which raises the concentration of radon inside a building[17].

Radiation exposure both inside and externally results from the natural radioactivity in building materials. However, the intrinsic radioactivity, which mostly affects the respiration system is mostly contributed by the progenies of radon and not by radon itself, which are released into the indoors from the building material like cement[18]. Radon progeny that assembles inside the lungs during respiration and irradiates the lung tissue, damages the cells and perhaps leads to lung cancer[19-22].

The goal of the survey is to, quantify and assess the radon concentrations in different geological settings. This information can further be used to locate sites for suitable construction material for new buildings as well as the identification of radon-prone areas. The rate of radon exhalation in soil samples has been quantified by numerous researchers all over the world. In the Hanumangarh district of Northern Rajasthan, the evaluation of exhalation rates of radon and thoron are quantified for the first time utilising Smart RnDuo, and their results are compared with those obtained from previous studies conducted in other regions of India.

## 2 Study Area

Hanumangarh district is located in the northern part of the state of Rajasthan in India. The study area stretches from 73° 47′ to 75° 32′ east longitude and 28° 46′ to 29° 57′ north latitude. Churu borders the district on the south, Sri Ganganagar on the west, Punjab on the north, and Haryana on the east. The geographic location of the study region and the sampling location are shown in Fig. 1. With a population of 1,774,692 and a vast area of 9656 km$^2$, it is notable from the standpoint of public health risk analysis. Both groundwater and surface water are the main sources of the water used for drinking. The Ghaggar River, the sole river in the area, is transient in nature. Nevertheless, the region's water requirements are met by the Indira Gandhi canal network, which was built to transport water from perennial Himalayan rivers flowing through Punjab to the desert parts of Rajasthan. When it comes to groundwater, it can be found in both limited aquifers and below the water table. Alluvium, which consists of sand, silt, clay, and gravel, is the main aquifer in

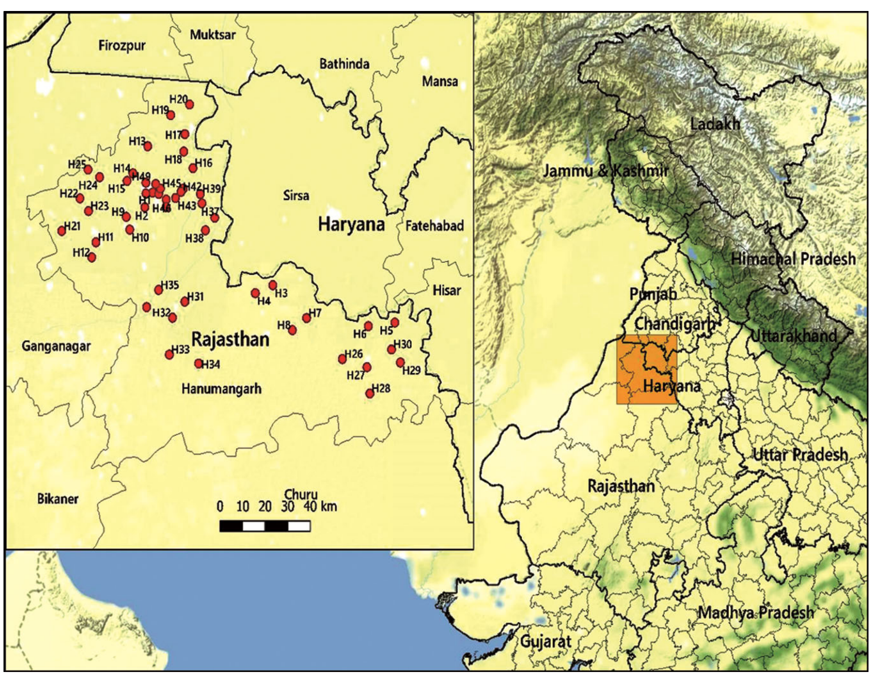


Fig. 1 — Map of Study Area: Hanumangarh district, Rajasthan, India.

the area. It has a thickness range of 100 to 400 metres. The Palana or Nagaur series of rocks, which are composed of mudstone, sandstone, and basal evaporations sequence, make up the bed rock beneath alluvium.

## 3 Methodology

Samples were taken from a variety of study area locations, as shown in the Fig. 1 and total 50 soil samples were collected from 25 distinct villages of Hanumangarh district. Also, the gamma level at each sample location was measured using a gamma survey metre (Polimaster PM-1405, Republic of Belarus) and the detector was positioned one metre above the ground. The Polimaster PM-1405 can monitor dose rates between 0.01 and 130Svh$^{-1}$ with a gamma energy response ranging from 0.05 to 3 MeV. A GPS map was used to record the coordinates, longitude, and latitude. The soil samples of 1000 gram weight were collected at each location from a depth of twenty centimetre from the soil profile, so as to acquire the refine sample. The acquired samples were pestled and filtered in order to maintain uniformity. The soil was baked in an oven to dry it out. To calculate exhalation rates of radon/thoron, a scintillation-based active detector called Smart RnDuo was used. It operates on the idea of detecting alpha particles that strike the scintillator and cause the ZnS:Ag coating to produce scintillations inside the cell. Using an integrated algorithm, the Photomultiplier Tube counts the scintillations and transforms them to radon/thoron concentration. In order to calibrate the detector, the detector is being taken to Bhabha Atomic Research Centre(BARC), Mumbai, India, once in a year.

The soil samples were placed in the mass exhalation chamber, which has a constant volume (540 cm$^3$) and a height of approximately 8.5 cm and is equipped with a detector, to quantify the exhalation rates (Smart RnDuo).It is based on the idea that the diffusion length of the radon is about one metre and that of thoron is about one centimetre, so only top layer is expected to contribute the thoron exhalation and the contribution of radon is expected from the whole sample due to considerable difference in their diffusion lengths. In addition, because thoron exhalation is a surface event, the thickness of the soil sample has no bearing on how large it is, making it a superior metric to describe thoron exhalation potential[25]. As a result, the design of the chamber will have no effect on the exhalation rates.

### 3.1 Radon mass exhalation rates

Since radon is a radioactive gas, we measure the mass exhalation rate of radon, which is the amount of radon that is released into the air per minute from a particular mass of soil matrix. The Smart RnDuo was used in diffusion mode to measure the rate at which radon mass exhaled. As depicted in Fig. 2, the soil samples were placed in the accumulation chamber, connected to the detector. To measure radon, gas was drawn into a scintillation chamber (153 cc), the radon gas was separated from its progeny and from thoron via “progeny filter” and “thoron discriminator”. Thoron discriminator's "diffusion time delay" prevents thoron from entering the detector. Radon estimation inside the detector is based on the scintillations produced by the alpha particles produced by the radon and its progenies inside the chamber of detector. According to the methodology, the detector was run continuously for twelve hours for every sample and 12 measurements were taken each of duration of 60 minutes. The exhalation for the sample was calculated using the standard formula and using the measured values.

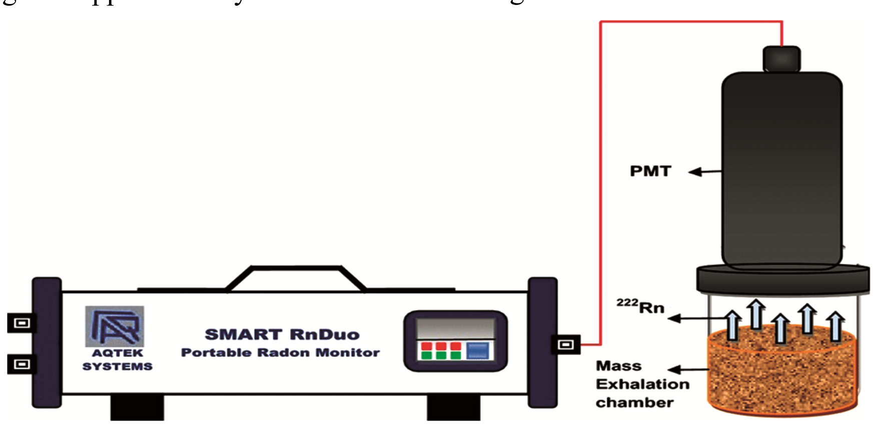


Fig. 2 — Setup for radon mass exhalation rates measurement (diffusion mode) in soil samples.

**3.2 Formulas used for quantification of radon exhalation rates in soil**

The data was calculated using the formula and the measurements of the detector and the graph was plotted between the concentration and the time (t) and slant of the graph was estimated by employing the least square fitting approach, as described in equation 1[26]. The equation 2 stated below was employed to calculate the radon mass exhalation rate of radon from the slope of the graph.

$$C(t) = (\frac{J_{Rm}M}{V})t + C_o \quad \ldots (1)$$

$$J_{Rm} = slope \times \frac{V}{M} \quad \ldots (2)$$

Where $J_{Rm}$ ($Bqkg^{-1}h^{-1}$) is the radon mass exhalation rate, $C_0$stand for the radon concentration ($Bqm^{-3}$) present in the chamber volume ($cm^3$) at t = 0, M represents a sample of dry soil's entire weight (kg) inside the chamber, t represents the measurement of time (h), and V is the effective volume (volume of detector + porous volume of sample ($Vp = Vs - \frac{M}{\rho_g}$)+ residual air volume of the mass exhalation chamber) ($m^3$).Where, $\rho_g$ is the sample's specific gravity which can be taken as 2.7 gm/cc for clay type soil sample.

**3.4 Thoron surface exhalation rates**

Thoron has a short diffusion length in the atmosphere, therefore its distribution in the accumulator is not uniform. So, an internal pump with a flow capacity of 0.7 $Lmin^{-1}$ was used and the detector was operated in "flow mode" to calculate the surface exhalation rate for thoron in soil samples (Fig. 3). The air that contains thoron gas (in the chamber) is circulated into the lucas cell by an internal pump in a closed loop after first passing through a "progeny filter." The alpha particles produced by the thoron and its offspring that have gathered inside the detector have been used to measure thoron activity. The measurement has been carried out for the course of four cycles of 15 minutes each to estimate the equilibrium thoron concentration ($C_T$) in the accumulation chamber. In the 15-minute cycle where the detector is operated on thoron mode, the sampling pump is maintained ON for the 5 minutes which gives the measure of the thoron and background, then there is a 5 minute delay to ensure the thoron's decay, and finally, 5 minutes of counting give the cycle's background counts. Following that, the thoron surface exhalation rates were computed using the measured value of $C_T$.

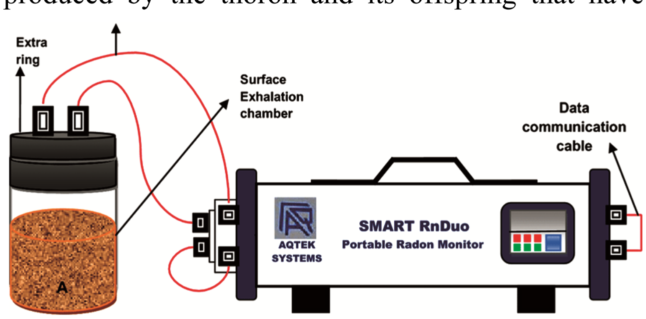


Fig. 3 — Setup for thoron surface exhalation rates measurement (flow mode) in soil samples.

**3.5 Formula used for quantification of thoron surface exhalation rates in soil**

The thoron surface exhalation rate has been calculated at the equilibrium using the following equation (3) (Radon handbook, BARC).

$$J_{Ts} = \frac{C_T V_T \lambda}{A} \quad \ldots (3)$$

Where,

$J_{Ts}$: the thoron surface exhalation rate of soil sample ($Bqm^{-2}h^{-1}$),

A: surface area of the chamber ($m^2$),

$V_T$: effective volume ($m^3$) (includes residual volume of chamber, internal volume of detector, volume of cylindrical pipes, residual volume of lid over the chamber),$C_T$ is the thoron build up concentration inside the chamber ($Bqm^{-3}$), and λ ($0.0126s^{-1}$ assuming half-life is 55s) is the decay constant of thoron.

## 4 Results and Discussion

In total 50 soil samples, the values of the radon mass exhalation rate has been found to stretch from 11.56 ± 0.62 $mBqKg^{-1}h^{-1}$ to 62.03 ± 3.49 $mBqKg^{-1}h^{-1}$ with mean value of 34.60 ± 1.37 $mBqKg^{-1}h^{-1}$ and thoron surface exhalation " rate varied from 3.09 ±0.39 to 9.56 ± 0.62 kBq $m^{-2}h^{-1}$ with mean value of 6.44 ±0.62 $kBqm^{-2}h^{-1}$ as shown in Table 1.

The frequency distribution shows that the radon mass exhalation rate of,4% soil samples (2)lying between 0 -13 $mBqkg^{-1}h^{-1}$, 14% soil samples (7) lying between 13-26 $mBqkg^{-1}h^{-1}$, 48% samples (24) lying between 26-39 $mBqkg^{-1}h^{-1}$ and for the rest of 34% samples (17) lie between39-65 $mBqkg^{-1}h^{-1}$ as shown in Fig. 4(a). It is depicted from the frequency distribution that the thoron surface exhalation rate of,

Table 1 — Radon mass/ thoron surface exhalation rates in studied area.

| Sample No. | Village | Geographical coordinates | Elevation (metres) | Gamma Level ($\mu Rh^{-1}$) | Radon mass exhalation rate ($J_{Rm}$) ($mBqkg^{-1}h^{-1}$) ± σ | Thoron surface exhalation rate ($J_{Ts}$) ($kBqm^{-2}h^{-1}$) ± σ |
|---|---|---|---|---|---|---|
| 1 | Hanumangarh. Junction | 74°18'08.13" | 165 | 0.121 | 30.72 ± 2.06 | 7.05 ± 0.55 |
| 2 | | 29°36'40.90" | | | 26.21 ± 1.98 | 6.45 ± 0.43 |
| 3 | Phephana | 74°54'58.94" | 164 | 0.164 | 12.23 ± 1.62 | 6.18 ± 0.51 |
| 4 | | 29°20'00.11" | | | 11.56 ± 1.23 | 6.35 ± 0.78 |
| 5 | Padampura | 74°55'36.45" | 163 | 0.175 | 32.83 ± 1.19 | 7.49 ± 0.59 |
| 6 | | 29°17'57.09" | | | 35.19 ± 1.24 | 8.51 ± 0.67 |
| 7 | Janania | 74°56'52.64" | 171 | 0.116 | 36.23 ± 2.10 | 8.38 ± 0.62 |
| 8 | | 29°20'09.99" | | | 37.76 ± 2.54 | 9.11 ± 0.54 |
| 9 | Jandawali | 74°13'11.33" | 170 | 0.138 | 40.76 ± 1.80 | 6.21 ± 0.56 |
| 10 | | 29°38'39.66" | | | 43.81 ± 1.92 | 7.33 ± 0.78 |
| 11 | Dabli Rathan | 74°11'27.56" | 162 | 0.164 | 36.53 ± 1.12 | 5.95 ± 0.49 |
| 12 | | 29°32'54.03" | | | 39.42 ± 1.34 | 6.98 ± 0.66 |
| 13 | Sangria | 74°27'52.64" | 165 | 0.157 | 29.59 ± 0.67 | 5.56 ± 0.48 |
| 14 | | 29°48'17.74" | | | 33.89 ± 0.94 | 7.43 ± 0.56 |
| 15 | Pirkamria | 74°28'06.69" | 160 | 0.158 | 40.30 ± 1.64 | 7.74 ± 0.55 |
| 16 | | 29°36'17.76" | | | 45.55 ± 1.76 | 8.84 ± 0.32 |
| 17 | Dhaba | 74°30'36.14" | 178 | 0.165 | 38.14 ± 1.99 | 5.34 ± 0.47 |
| 18 | | 29°52'35.15" | | | 40.15 ± 2.14 | 6.15 ± 0.57 |
| 19 | Bhagat Pura | 74°27'14.68" | 169 | 0.172 | 42.73 ± 0.89 | 5.39 ± 0.46 |
| 20 | | 29°50'56.12" | | | 41.82 ± 0.94 | 6.23 ± 0.32 |
| 21 | Nagrana | 74°25'29.39" | 166 | 0.11 | 62.03 ± 1.57 | 5.93 ± 0.50 |
| 22 | | 29°39'44.14' | | | 61.11 ± 1.69 | 6.32 ± 0.72 |
| 23 | Goluwala | 74°07'22.19" | 167 | 0.171 | 46.05 ± 1 | 6.25 ± 0.52 |
| 24 | | 29°37'49.84' | | | 48.10 ± 1.12 | 7.78 ± 0.64 |
| 25 | Tibbi | 74°30'07.41" | 166 | 0.123 | 29.75 ± 1.32 | 3.99 ± 0.41 |
| 26 | | 29°33'13.06" | | | 30.14 ± 1.64 | 4.25 ± 0.55 |
| 27 | Panniwala | 74°25'50.47" | 175 | 0.124 | 45.84 ± 1.42 | 8.57 ± 0.58 |
| 28 | | 29°36'26.14" | | | 44.89 ± 1.67 | 8.98 ± 0.77 |
| 29 | Kharliya | 73°59'47.54" | 169 | 0.138 | 44.48 ± 3.49 | 6 ± 0.21 |
| 30 | | 29°33'39.30' | | | 45.35 ± 2.87 | 6.71 ± 0.34 |
| 31 | Hansliya | 74°02'39.49" | 164 | 0.78 | 37.81 ± 1.05 | 6.8 ± 0.53 |
| 32 | | 29°34'29.17' | | | 40.42 ± 1.23 | 6.9 ± 0.61 |
| 33 | 25 DWD | 74°19'41.40" | 169 | 0.174 | 20.25 ± 1.06 | 5.54 ± 0.49 |
| 34 | | 29°15'53.70" | | | 22.54 ± 1.88 | 6.61 ± 0.55 |
| 35 | Rawatsar | 74°24'31.98" | 167 | 0.135 | 14.24 ± 0.62 | 3.34 ± 0.39 |
| 36 | | 29°15'51.57" | | | 15.33 ± 0.71 | 3.45 ± 0.55 |
| 37 | Hanumangarh. Town | 74°53'60.13" | 175 | 0.161 | 27.80 ± 0.83 | 7.32 ± 0.53 |
| 38 | | 29°32'33.09" | | | 34.93 ± 1.76 | 9.56 ± 1.21 |
| 39 | Dholpalia | 75°11'17.37" | 181 | 0.164 | 31.67 ± 0.91 | 6.37 ± 0.55 |
| 40 | | 29°02'51.45" | | | 30.88 ± 0.88 | 7.06 ± 0.77 |
| 41 | Bhadra | 75°11'17.37" | 183 | 0.134 | 23.04 ± 0.78 | 3.09 ± 0.43 |
| 42 | | 29°06'57.24" | | | 27.88 ± 1.15 | 4.23 ± 0.75 |
| 43 | 2 KBM | 74°22'22.61" | 188 | 0.132 | 17.62 ± 0.90 | 3.67 ± 0.41 |
| 44 | | 29°15'02.06" | | | 19.71 ± 1.16 | 5.15 ± 0.95 |
| 45 | Jodkiyan | 74°17'07.91" | 179 | 0.146 | 35.25 ± 1.46 | 6.28 ± 0.51 |
| 46 | | 29°39'30.05" | | | 38.96 ± 1.86 | 7.21 ± 0.94 |
| 47 | Jogiwala | 75°16'21.07" | 167 | 0.144 | 34.54 ± 1.78 | 6.37 ± 0.51 |
| 48 | | 29°06'25.92" | | | 37.97 ± 1.99 | 7.67 ± 0.77 |
| 49 | Pillibanga | 74°04'41.14" | 162 | 0.168 | 31.34 ± 0.88 | 5.9 ± 0.5 |
| 50 | | 29°39'09.41" | | | 34.78 ± 1.11 | 6.08 ± 0.89 |

10% soil samples (5) lie between 0-4 $kBqm^{-2}h^{-1}$, for 76% soil samples (38) lie between 4-8 $kBqm^{-2}h^{-1}$ and rest of 14% soil samples (7) lie between 8-12 $kBqm^{-2}h^{-1}$ as shown in Fig. 4(b). These values are lower than the worldwide mean value of 57 $mBqKg^{-1}h^{-1}$ for radon mass exhalation rate and higher than the worldwide value of 3.6 $kBqm^{-2}h^{-1}$ for thoron surface exhalation rate[27].

**4.1 Correlation between indoor radon/thoron concentration with radon/thoron exhalation rates**

The relationship between radon exhalation rates and indoor radon concentration has been studied. The linear fit in Fig. 5(a) has an $R^2$ value of 0.13 and a Pearson's r value (which measures linear dependency) of 0.36, both of which point to an extremely tenuous relationship between the two variables. The analysis was also performed to investigate the relationship between indoor thoron concentration and thoron exhalation rates. The linear fit in Fig. 5(b) has an $R^2$ value of 0.01 and a Pearson r value of 0.104, both of which indicate a very poor relationship between the two variables.

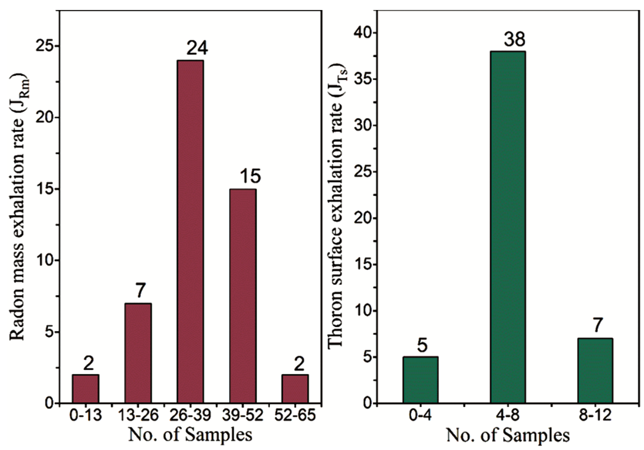


Fig. 4 — Frequency Distribution for (a) radon mass (b) thoron surface exhalation in soil samples.

The radon mass exhalation rates in soil samples have lower values than 57 $mBqkg^{-1}h^{-1}$ and thoron surface exhalation rates were higher than 3.6 $kBqm^{-2}h^{-1}$ as recommended by UNSCEAR, 2000, in Hanumangarh district of Northern Rajasthan. The very high thorium levels in these soil samples may be the cause of the intense thoron concentration and surface exhalation rate. It closely resembles the outcomes of the higher thorium in the northern part of India which was shown in the radiation profile map of India[28-30].

**4.2 Comparison with similar investigations performed by other authors**

The variation in exhalation rates may be due to certain factors like geology of study area, radon emanation factor and porosity of soil samples as reported by BK Sahoo[31]. The findings of the research area have been contrasted with those of similar studies carried out in other regions of India as shown in Table 2. These are slightly higher than the studies carried out in Tarantaran and Amritsar districts of Punjab[32], Shiwalik Himalayas of Jammu and Kashmir[33]. These values are lower than uranium mineralization area Una and Hamirpur districts of Himachal Pradesh[34], Aravali Hills[35], Cauvery River sediments[36], plain area Faridabad, Haryana[37], worldwide average value of 57 $mBqkg^{-1}$ $h^{-1}$[27].

Thoron exhalation varies depending on the geography and geology of the soil sample location. The effects of a greater level of thorium are often

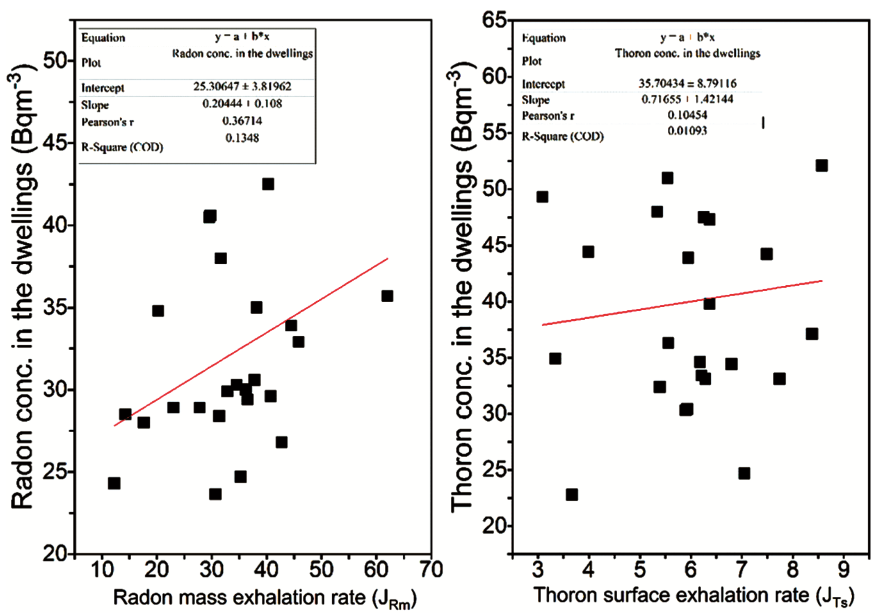


Fig. 5 — Correlation between (a) Radon concentration in the dwellings with radon mass exhalation rates in soil samples (b) thoron concentration in the dwellings with thoron surface exhalation rates in soil.

Table 2 — Comparison with other, related research from other areas.

| S. No. | Place | Radon mass exhalation rates ($J_{Rm}$) ($mBq\ kg^{-1}\ h^{-1}$) ± σ | Thoron surface exhalation rates ($J_{Ts}$) ($Bq\ m^{-2}\ s^{-1}$) ± σ | Reference |
|---|---|---|---|---|
| 1. | Tarn Taran | 23 ± 5 | 1531 ± 1503 | 32 |
| 2. | Amritsar | 20 ± 7 | 664 ± 237 | 32 |
| 3. | Jammu and Kashmir | 8 ± 1 to 62 ± 3 | 295 to 3628 | 33 |
| 4. | Shiwalik Himalayas of J& K | 7 to 48 | 123 to 2606 | 32 |
| 6. | Aravali hills | 23 to 50 with average of 35 | - | 35 |
| 7. | Una and Hamirpur Himachal Pradesh | 39 to 91 with average of 60 | - | 34 |
| 9. | Cauvery river sediments | 45 to 333 | - | 36 |
| 10. | Faridabad, Haryana | 12 ± 1 to 62 ± 4 with average of 31 ± 12 | | 37 |
| 12. | Kalpakkam, Tamilnadu | - | 942 to 7720 | 38 |
| 13. | Hanumangarh district | 11.56 ± 0.62 to 62.03 ± 3.49 with average of 34.60 ± 1.37 | 3.09 ± 0.39 to 9.56 ± 0.62 with average of 6.44 ± 0.62 | Present study |

strongly associated with the effects of a higher level of thoron in the northern part of India, which arose in the radiation profile guide of India, because rocks have a higher thorium content than other materials since the earth's origin. The findings of the present study are comparable to those of studies conducted in the other Indian regions listed above. The thoron surface exhalation rates are higher than Taran Taran and Amritsar districts of Punjab[32], Himalayas of Jammu and Kashmir[33], Shiwalik Himalayas of Jammu and Kashmir, Kalpakkam, Tamilnadu[38] and worldwide mean value of 5700 $Bqm^{-2}h^{-1}$.

## 5 Conclusions

In comparison to the worldwide average of 57 $mBq\ kg^{-1}\ h^{-1}$, the mass exhalation rates of radon have been lower. The exhalation rates near the surface that is of thoron were higher than the global average of 3.6 $kBq\ m^{-2}\ h^{-1}$. The variance in exhalation rates may be caused by the terrain, radon emanation factor, different geological soil sample sites, and soil porosity. The increased concentration of thorium-rich materials in the rocks of northern India may be the cause of the elevated thoron surface exhalation rates.

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