# **Quantifcation of 222Rn/ 220Rn exhalation rates from soil samples of Champawat region in Kumaun Himalaya, India**

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#### **Abstract**

The present study is carried out in selected locations of the Champawat district in the lesser Kumaun Himalayan belt of Uttarakhand, India. The Scintillation detector-based portable Smart RnDuo monitor was used to estimate exhalation rates of  $222$ Rn and  $220$ Rn. The levels of gamma dose rates were also measured using a portable radiation survey meter at each of the selected locations. The mass exhalation rate for <sup>222</sup>Rn varies between  $7.8 \pm 0.1$  and  $107.3 \pm 2.1$  mBq kg<sup>-1</sup> h<sup>-1</sup> with an average of 38.9 $\pm$ 18.9 mBq kg<sup>-1</sup> h<sup>-1</sup>. In contrast, the <sup>220</sup>Rn surface exhalation rate ranges from 0.7 $\pm$ 0.5 to 14.3 $\pm$ 0.7 Bq m<sup>-2</sup> s<sup>-1</sup>with an average  $6.2 \pm 3$  Bq m<sup>-2</sup> s<sup>-1</sup> and emanation rate was found to range 88.4 and 1703.7 mBq kg<sup>-1</sup> s<sup>-1</sup> with an average  $739 \pm 356.7$  mBq kg<sup>-1</sup> s<sup>-1</sup>. The gamma dose rate was found to vary from  $0.08 \pm 0.01$  to  $0.30 \pm 0.03$   $\mu$ Sv h<sup>-1</sup> with an average of  $0.18 \pm 0.06$   $\mu$ Sv h<sup>-1</sup> in the region.

**Keywords** Natural background radiation · Smart RnDuo monitor · Exhalation rates · Gamma dose rate

# **Introduction**

Soil, water and air are diferent phases contributing towards the distribution of natural background radioactivity to the ambient environment of inhabitants with abundantly available radioactive minerals as the significant source term  $[1-8]$  $[1-8]$ . Natural background radiation dose values are reported in a number of studies performed in diferent parts of the globe [\[9](#page-9-2)[–12](#page-9-3)]. Radon (<sup>222</sup>Rn) and its isotope thoron (<sup>220</sup>Rn) are the disintegrated products of radium ( $^{226}$ Ra) and ( $^{232}$ Th), respectively. These gaseous ( $^{222}$ Rn and  $^{220}$ Rn) are found throughout the earth's crust in varying concentrations. It has been found that  $^{222}$ Rn,  $^{220}$ Rn and their decay products contribute more

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than 50% of the total dose to the inhabitants from natural background radiation sources [[13\]](#page-9-4).

The amount of radon and thoron in the indoor air is directly related to the presence of the radium  $(^{226}Ra)$  and thorium  $(^{232}Th)$  radionuclides in soil. Rate of radon exhalation from the soil was observed to be positively linked with indoor radon concentration and radium content in soil  $[14–16]$  $[14–16]$  $[14–16]$  $[14–16]$ . Radon and thoron gases are formed by the disintegration of radium and thorium, respectively in the earth's crust. These radioactive gases come to the upper surface of the atmosphere through exhalation and emanation processes. Emanation is a process by which  $^{222}$ Rn atoms escape from the solid mineral grains to the air-flled pores in the soil matrix. In contrast, exhalation is the process by which radon gas is transported from air-flled pores to the earth's atmosphere. The primary contributors to the levels of radon  $(^{222}Rn)/th$ oron  $(^{220}Rn)$  concentration in the indoor air and outdoor atmosphere are both the source terms, i.e., exhalation and emanation from soil and building materials [[17–](#page-9-7)[20](#page-9-8)].

Radon/thoron is easily transported from the grain particles to the soil pore space (i.e., emanation) and from airflled pores to the environment or indoor air (exhalation). Difusion and advection processes can move radon from the pore space to the soil-air interface. However, in the vast majority of cases, radon is transported through difusion.

The radon is released into the atmosphere after emanation and transportation within the soil [[21](#page-9-9)].

There are several factors which infuence radon emanation and exhalation from soil or rock, including radium distribution, grain size, porosity, moisture content, and so on. All of these have a signifcant impact on the exhalation and emanation process [[22](#page-9-10)[–25\]](#page-9-11). Radium distribution and its relationship to radon exhalation rate and soil-gas radon concentrations are likely to vary in various formations and rock types [[26\]](#page-9-12). Moreover, the external radiation that arises due to exhalation from terrestrial radioactive residues accounts for around 20% of the natural radiation dose [[27](#page-9-13)]. As radon gas is inhaled, densely ionising alpha particles emitted from short-lived radon decay products  $(^{218}Po$  and  $^{214}Po$ ) interact with biological tissues causing lungs damage [\[19\]](#page-9-14). If the concentration of radon, thoron and its daughter products in the closed spaces such as houses, large buildings, and mines exceeds the threshold, it may pose a radiation hazard to the occupants [[28–](#page-9-15)[30](#page-9-16)].

In the present study, radon mass exhalation rate and thoron surface exhalation rate were measured in Champawat region of Kumaun Himalaya, Uttarakhand, India. The ambient gamma dose rates were measured using a portable radiation survey meter at each sampling location. The results of the measurements of exhalation rates in the current work would provide an essential link with indoor radon, thoron and their progeny concentrations in the region. The results of the present study may be helpful in future studies in the feld of environmental radioactivity.

#### **Study area**

The present study was conducted in the Champawat region which lies in the vicinity of the Kumaun Himalaya, Uttarakhand, India. The Champawat district is located in the southern part of Uttarakhand state (28°N to 29.5° N and 79.7° 34′ E to 80.3° E) and shares an international border with Nepal in the East (Fig. [1\)](#page-1-0). Champawat district covers a total area of  $1765.8 \text{ Km}^2$  with an average altitude of

<span id="page-1-0"></span>



1670 m above sea level. The Kali River divide Uttarakhand and Nepal in the East. The Ladhiya and Lohawati Rivers are the major drainage system in the area. The route from south to north traverses across Bhabhar Formation, Siwalik Group of rocks, Bhimtal Quartzites and Almora Crystallines. Signifcant tectonic plates such as the Himalayan Frontal Thrust (HFT), Main Boundary Thrust (MBT), and South Almora Thrust (SAT) distinguish the geological formations Current-bedded quartzite with associated volcanic, mica-talc schist, limestone, conglomerate, slate, quartzite, granodiorite, augen gneiss, migmatite, and granite gneiss are found in various parts of the Champawat district. Champawat area is characterized by various units viz. Alluvium plain, Siwalik Group, Outer Lesser Himalaya and the Almora Crystalline unit [[31,](#page-9-17) [32\]](#page-9-18).

# **Materials and methodology**

# **Measurements of 222Rn/ 220Rn exhalation rates**

 $222$ Rn and  $220$ Rn exhalation rates from soil were measured using a scintillation detector based Smart RnDuo radon monitor. The Smart RnDuo is a real-time portable radon monitor. The alpha scintillations from radon and its decay products are counted via a photo multiplier tube (PMT) and the associated counting electronics. The Smart RnDuo has a quick reaction time and operates in a permanent, maintenance-free mode with long-term calibration stability and suitable for radon monitoring in both indoor as well as outdoor conditions. This equipment can be used to test radon levels in soil, water, and the Naturally Occurring Radioactive Materials (NORMs). The detection limit of Smart RnDuo ranges from 8 Bq m<sup>-3</sup> to 50 MBq m<sup>-3</sup> [\[33\]](#page-9-19). The investigations for the present study were carried out in the diferent areas situated in the entire Champawat district of Kumaun Himalaya, India. The detailed description of measurement procedures of surface and mass exhalation rates is given below. The gamma dose rates were also measured using a radiation survey meter manufactured by ATOMTEX (Belarus) at each sampling location.

#### **Measurements of thoron surface exhalation rates**

The Smart RnDuo monitor was employed to measure thoron surface exhalation rates. Thoron is not uniformly distributed in the accumulation chamber on account of its very short difusion length. Therefore, the Smart RnDuo monitor was operated in flow mode to estimate surface exhalation of thoron. Smart RnDuo has an in-built function for the flow mode operation. It sets the pump ON automatically for 5 min in 15 min cycle or 15 min in 1-h cycle, respectively. To measure  $^{220}$ Rn surface exhalation rates, 15 min cycles

were taken and the air volume in the chamber was kept to a minimum to ensure proper air mixing inside the closed-loop. The measurement procedure of thoron surface exhalation is shown in Fig. [2](#page-2-0). The detailed measurement principle can found in the literature [\[34](#page-9-20), [35\]](#page-10-0). For calculating the steadystate 220Rn concentration (Bq m−3)**,** the averages of four values were considered for the estimation. The Eq. [\(1](#page-2-1)) is used to calculate the surface exhalation rate  $(J<sub>S</sub>)$ 

<span id="page-2-1"></span>
$$
J_S = C_T V \lambda / A \tag{1}
$$

where,  $C_T$  represents the average concentration of thoron inside the accumulation chamber (Bq m<sup>-3</sup>), *V*,  $\lambda$  and *A* are the residual air volume  $(m<sup>3</sup>)$ , effective decay constant for  $^{220}$ Rn (h<sup>-1</sup>) and the surface area of the chamber (m<sup>2</sup>), respectively. The mass emanation rate of  $^{220}$ Rn from soil can be determined using Eq. [\(2](#page-2-2)) [\[36](#page-10-1)]

<span id="page-2-2"></span>
$$
J = [J_S/(\rho \times L_S)]\tanh(Z_S/L_S)
$$
 (2)

where,  $J_s$  = surface exhalation rate,  $Z_s$  = sample height,  $Ls =$ diffusion length of <sup>220</sup>Rn within soil and  $\rho =$  density of soil samples. The ratio  $Z_{\gamma}/L_{s}$  is greater than unity since the diffusion of thoron  $(L<sub>c</sub>)$  is about 1 cm [[37\]](#page-10-2). Using this approximation, Eq. [\(2](#page-2-2)) can be represented as

<span id="page-2-3"></span>
$$
J = J_S/(\rho \times L_S) \tag{3}
$$

The Eq. ([3\)](#page-2-3) is used to estimate thoron mass exhalation rate.

# **Measurement of mass exhalation rate of 222Rn**

The soil samples (each sample  $\approx 1$  kg) were collected from the ground at a depth of 30 cm and dried under the sunlight. The roots, stones, peebles were removed from the ground surface before collecting the samples. The measurements of radon mass exhalation rates in the collected samples were carried out in the laboratory.



**Exhalation Chamber** 

<span id="page-2-0"></span>**Fig. 2** Schematic diagram in Flow mode of Smart RnDuo for surface exhalation measurement



**Exhalation Chamber** 

<span id="page-3-0"></span>**Fig. 3** Schematic diagram in difusion mode for Mass Exhalation Using Smart RnDuo

The schematic diagram of the measurement procedure is shown in Fig. [3](#page-3-0). The Smart RnDuo monitor is connected to a close cylindrical and leakage-free accumulation chamber made of stainless steel. For  $222$ Rn measurement, the Smart RnDuo monitor was programmed to run for 24 h on a 60-min cycle. The monitor is operated in diffusion mode to avoid the thoron interferences since thoron has a very short diffusion length as compared to that of radon. The detailed explanation of the Smart RnDuo as well as the measuring technique is given elsewhere [[33](#page-9-19)]. The growth of radon concentration  $C_t$  at time  $t$  (Bq m<sup>-3</sup>) in the accumulated chamber is given by following Eq. ([4\)](#page-3-1)

$$
C_t = \left(J_m M / V \lambda_e\right) \left[1 - e^{-\lambda e t}\right] + C_0 e^{-\lambda e t} \tag{4}
$$

where,  $C_t$ ,  $J_m$ ,  $M$ ,  $V$ , and  $\lambda_e$  represent the concentration of 222Rn at any time *t* (Bq m−3), mass exhalation rate (Bq  $kg^{-1} h^{-1}$ ), sample mass (kg), volume of the chamber plus scintillation cell  $(m^3)$ , effective decay constant  $(h^{-1})$  which is the summation of leakage rates of  $^{222}$ Rn and decay constant of the accumulator if it exits. At  $t=0$ ,  $C_0$  represents initial  $222$ Rn concentration in the chamber. The data is further linearly ftted as per the Eq. ([4\)](#page-3-1) to obtain mass exhalation rate  $(J_m)$ .

In linear approximation, Eq. [\(4\)](#page-3-1) can be transformed into the following Eq. ([5\)](#page-3-2)

$$
C_t = (J_m M/V)t + C_0 \tag{5}
$$

The graph was plotted between radon concentration data (Bq m<sup>-3</sup>) with time (in hour) and it gives the wellknown exponential growth curve of radon accumulation as shown in Fig. [4](#page-3-3) [[38,](#page-10-3) [39](#page-10-4)].



<span id="page-3-3"></span>**Fig. 4** Growth curve of Accumulated Radon

#### **Measurements of gamma dose rate**

The Radiation Survey Meter manufactured by ATOM-TEX (Belarus) was used to measure the gamma dose rates. Gamma dose rate were measured 1 m above the ground surface at all sampling locations. The gamma dose rate survey may be hepful in the estimation of terrestrial radiation in the study area.

### **Result and discussion**

# **222Rn mass exhalation rate from the soil samples**

<span id="page-3-1"></span>Table [1](#page-4-0) represents the measured values of gamma dose rate  $(\mu Sv h^{-1})$ , <sup>222</sup>Rn mass exhalation rate (mBq kg<sup>-1</sup> h<sup>-1</sup>), <sup>220</sup>Rn Surface exhalation rate (Bq m<sup>-2</sup> s<sup>-1</sup>) and <sup>220</sup>Rn emanation rate (mBq kg<sup>-1</sup> s<sup>-1</sup>) for collected samples and statistical parameters (minima, maxima, mean and standard deviation.

<span id="page-3-2"></span>The values of  $222$ Rn mass exhalation rate varied from 7.8 ± 0.1 to 107.3 ± 2.1 mBq kg<sup>-1</sup> h<sup>-1</sup>with an average of  $38.9 \pm 18.9$  mBq kg<sup>-1</sup> h<sup>-1</sup>. Topological studies show that the high exhalation rate from the soil matrix causes a high level of terrestrial radiation exposure [\[40](#page-10-5)]. The higher value of  $^{222}$ Rn mass exhalation rate can be attributed to the higher  $^{226}$ Ra content in the soil samples. The high values of mass exhalation rates were found for samples C10, C13, C41, C42, C43 and C44. These specifc high values are caused by high radium content in the porous soil matrix. The low values of mass exhalation rates correspond to samples C2 and C4. The diferent geological formations and soil composition tend to obtain diferent exhalation rates determined by the abundance of parent source term [[41](#page-10-6)]. The formation alongside the main boundary thrust (MBT) like sand-stone bedrock and mudstone is also

<span id="page-4-0"></span>

responsible for such vast variation for a small region [[42](#page-10-7)]. The Fig. [5](#page-5-0) shows the statistical distribution of  $^{222}$ Rn mass exhalation rates of the samples.

The Box plot on the left shows that the mean is about 39 mBq kg<sup>-1</sup> h<sup>-1</sup> and the median is about 35 mBq kg<sup>-1</sup> h<sup>-1</sup>. The measured  $^{222}$ Rn mass exhalation rate of 25% and 75% of the samples is below and above 30 mBq kg<sup>-1</sup> h<sup>-1</sup>, respectively (Fig. [5\)](#page-5-0). The mean occurs in the third quartile that shows a higher mean value than the median value. The right side of the plot shows the kernel density of the data and the statistical distribution is approximated to a normal distribution. However, the dataset has a negative skewness because large numbers of the data points are lying beyond the central area of the kernel density. This indicates a wide distribution of  $^{222}$ Rn mass exhalation rate over the area.



Mass Exhalation Rate (mBq/Kg/h)

<span id="page-5-0"></span>Fig. 5 Box plot representing the statistics of mass exhalation data surface exhalation rate.

<span id="page-5-1"></span>**Table 2** Comparision of radon mass exhalation rates and thoron surface exhalation rates with previous studies from Northern India

# **220Rn surface exhalation rate and emanation rate**

The measured values of  $^{220}$ Rn surface exhalation rate (Bq  $m^{-2}$  s<sup>-1</sup>) and emanation rate (mBq kg<sup>-1</sup> s<sup>-1</sup>) along with their statistical parameters (minima, maxima, mean and standard deviation), are shown in Table [1](#page-4-0). The measured levels of surface exhalation of  $^{220}$ Rn vary from  $0.7 \pm 0.5$  and  $14.3 \pm 0.7$  Bq m<sup>-2</sup> s<sup>-1</sup> with an average of  $6.2 \pm 3$  Bq m<sup>-2</sup> s<sup>-1</sup> the diferent locations of Champawat. The higher levels of  $220$ Rn surface exhalation rate might be attributed to the higher concentration of thorium content in the soil profles. The results due to higher levels of 220Rn surface exhalation are comparable with the other studies in the Northern part of India as shown in Table [2.](#page-5-1) There were 44 soil samples collected from diferent locations in the investigated region, among them 52% samples indicate higher surface exhalation rate than the average value  $6.2 \pm 3$  Bq m<sup>-2</sup> s<sup>-1</sup>. The thoron emanation rate was to be found between 88.4 and 1703.7 mBq  $kg^{-1} s^{-1}$  with an average  $739 \pm 356.7$  mBq kg<sup>-1</sup> s<sup>-1</sup>. Different diffusion lengths of thoron cause variation in mass emanation rate. The difusion length depends upon the porosity of the soil matrix. Figure [6](#page-6-0) shows the Whisker's plot of  $^{220}$ Rn surface exhalation rate which indicates the statistical summary of the data. The median is slightly higher than the mean but can be approximated equally. This is the characteristic of a normal distribution with a fne goodness-of-ft but with some skewness. The right side of the plot shows a good approximation of the normal distribution for the dataset. Thoron surface exhalation rate of 25% of the samples was observed below 4 Bq m<sup>-2</sup> s<sup>-1</sup>. Figure [7](#page-6-1) shows a Whisker's diagram of  $220$ Rn emanation rate dataset. The mean of the dataset occurs in the second quartile of the Whisker's box, indicating that the median is higher than the mean. The emanation rate of 25% of the samples was found below 500 mBq  $kg^{-1} s^{-1}$ . The right side of the plot shows the kernel density of the dataset which shows that the whole data follows normal distribution like mass exhalation rate and





<span id="page-6-0"></span>**Fig. 6** Box plot representing the statistics of surface exhalation data



<span id="page-6-1"></span>**Fig. 7** Box plot representing the statistics of mass emanation data

# **Gamma dose rates**

The Gamma dose rates were measured at the same locations of study area from where the soil samples were collected and the measured values are shown in Table [1.](#page-4-0) The Gamma dose rate varies from  $0.08 \pm 0.01$  to  $0.30 \pm 0.03$   $\mu$ Sv h<sup>-1</sup> with an average  $0.18 \pm 0.06$  μSv h<sup>-1</sup>. Gamma dose rate gives a background radiation level due to all types of radiation sources, i.e., terrestrial and cosmic radiation.

# **Spatial distribution of 222Rn exhalation rate and 220Rn exhalation rate**

Figure [8](#page-7-0) shows the spatial distribution of <sup>222</sup>Rn mass and  $220$ Rn surface exhalation rate of the soil samples taken from the study area i.e., Champawat district of Uttarakhand State of India. As the dataset is pointwise located on the map of Champawat, the neighboring values were determined using inverse distance weightage approximation. The spatial distribution is then denoted with the colour-coded contouring intervals. This method does not provide the actual values on neighboring locations but gives a general idea of the approximate values on the locations from where the samples were not taken. In Fig. [8a](#page-7-0) the soil samples taken from locations C10, C42 and C43 show the higher values of  $^{222}$ Rn mass exhalation rate and the region in between shows the lower values (below 20 mBq kg<sup>-1</sup> h<sup>-1</sup>). Figure [8](#page-7-0)b shows that most of the region occurs at high surface exhalation rates and locations C2, C10, C18 and C35 have the highest values of  $J_s$  (i.e., 10.5–14.3 Bq m<sup>-2</sup> s<sup>-1</sup>). This indicates the possibility of a high abundance of thorium over the region.

### **Correlation study**

The Fig. [9](#page-8-0) shows a correlation matrix indicating the mutual correlations of gamma dose rate, <sup>222</sup>Rn mass exhalation rate,  $^{220}$ Rn surface exhalation rate and  $^{220}$ Rn emanation rate. The radon mass exhalation rate in the soil is plotted against thoron surface exhalation rates. A weak positive correlation with Pearson's value 0.10 was observed between radon mass exhalation rate and thoron surface exhalation rate. A signifcant correlation was found between the gamma dose rate and the radon mass exhalation rate with Pearson's value of 0.51. No signifcant correlation was observed between gamma dose rate and thoron surface exhalation rate with Pearson's value of 0.22. A strong positive correlation was found between emanation rate and surface exhalation rate of thoron with Pearson's value of 1. It indicates an obvious and signifcant relationship between the surface exhalation rate and emanation rate of thoron [[40,](#page-10-5) [41](#page-10-6)]. Emanation rates are showing the same trends of correlation with other entities, same as surface exhalation rate. The diagonal graphs show the histograms over the datasets, which provides the statistical distribution of the datasets.

#### **Comparison with previous studies**

Table [2](#page-5-1) represents values of radon mass exhalation rates and thoron surface exhalation rates previously reported in diferent parts of northern India. Similarly, Table [3](#page-8-1) shows the values of radon mass exhalation rates and thoron surface exhalation rates in diferent regions of the globe. The average value of  $^{222}$ Rn mass exhalation rate estimated in the investigated region is comparable with the recent studies, as shown in Tables [2](#page-5-1) and [3](#page-8-1). The  $^{220}$ Rn surface exhalation rate is found to be signifcantly higher than the values reported in other studies.



<span id="page-7-0"></span>**Fig. 8** Spatial distribution of  $\mathbf{a}^{222}$ Rn exhalation rate and  $\mathbf{b}^{220}$ Rn exhalation rate

# **Conclusions**

The study shows that the exhalation rates of  $222$ Rn and  $220$ Rn in the soil are controlled by several factors like porosity, geological features, and minerals present in the soil. However, Main Boundary Thrust (MBT) and local faults might have played a signifcant role in controlling exhalation and emanation rates in the study area. The average gamma dose rate was found  $0.18 \pm 0.06$  µSv h<sup>-1</sup>. The average mass exhalation rate in the study area was found to be  $38.9 \pm 18.9$  mBq kg<sup>-1</sup> h<sup>-1</sup> which is less than the global average 57 mBq kg−1 h−1. The observed variation may depend on the uranium mineralization in the study area. The high surface exhalation rate indicating rich thorium content in

the study area has an average of  $6.2 \pm 3$  Bq m<sup>-2</sup> s<sup>-1</sup>. A weak positive correlation observed between mass exhalation rate and surface exhalation rate can be linked to shorter half-life and difusion lenght for thoron. Most of the samples have shown higher surface exhalation rate than the global average value 1 Bq m<sup>-2</sup> s<sup>-1</sup>, which is in close agreement with more elevated surface exhalation rate in northern part of India (Table  $2$ ). A strong correlation coefficient between the thoron emanation and thoron surface exhalation rate shows that the exhalation rate depends on emanation. The results of the present study will be helpful to substantiate the measurements of radon, thoron and progeny levels in the study area.



<span id="page-8-0"></span>**Fig. 9** Matrix plot representing Pearson correlations of the statistical data

<span id="page-8-1"></span>**Table 3** Comparision of radon mass exhalation rates and thoron surface exhalation rates with some worldwide studies



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#### **Declarations**

**Conflict of interest** The authors declare that they have no confict of interest in this manuscript.

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