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# Estimation of terrestrial radionuclide concentration and effect of soil parameters on exhalation and emanation rate of radon



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#### ARTICLE INFO

### ABSTRACT

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Keywords: <sup>226</sup>Ra <sup>232</sup>Th <sup>40</sup>K Exhalation and emanation rate Soil parameters The soil samples collected from the different locations of the Jaipur and Ajmer districts of Rajasthan have been analysed for <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K content using gamma spectrometry. The average concentration of the <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K in soil samples comes out to be 69, 55 and 884 Bq kg<sup>-1</sup>. The Emanation and Exhalation rate of the <sup>222</sup>Rn, <sup>220</sup>Rn from the collected soil samples have been measured. As the <sup>222</sup>Rn and <sup>220</sup>Rn originates from the solid grains of the medium and migrate through its pore space, it is expected to get affected by various soil parameters. An attempt has been made to see the effect of physical soil parameters on the Exhalation and Emanation rate of the <sup>222</sup>Rn, <sup>220</sup>Rn meanation and migration through the medium.

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#### 1. Introduction

Uranium, Thorium and their decay product radionuclides are predominate part of the environmental radioactivity. Due to the presence in all three natural decay series, high mobility in the natural environment and bone seeking properties of radium, it require particular attention (Fesenko et al., 2009). The concentration of the natural radionuclides in the soil depends mainly upon the parent rock properties and also on the soil parameters. Soil is formed from the rocks due to various environmental phenomenon such as weathering, change in temperature etc. The weathering of the rocks causes the radionuclides to deposits in the soil or to reach the air and water medium (Iyengar, 1990).

The rocks and soil under or surrounding the homes are the main sources of indoor radon and thoron (Nazaroff et al., 1988a). Therefore indoor concentration of radon, thoron depends upon their parent radionuclide concentration in the soil or rock and on the access of the radon/ thoron gases to indoor dwellings. The second largest contribution of the radon and thoron in the indoor environment is the building material used in the construction of the dwellings (Nazaroff et al., 1988a; Stranden, 1988; UNSCEAR, 2000; Nazaroff, 1992). Other sources of the indoor radon and thoron are the groundwater, natural gases and outdoor air but the contribution of these three is negligible for an average home (UNSCEAR, 2000; Nero, 1988; Nazaroff et al., 1988b).

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The soil and the building material consist of the solid and porus fraction. The solid fraction consists mainly of mineral grain and organic matter but the porus fraction usually consists of the water and gas. The radon and thoron concentration in the rock or soil present around the dwellings is 10<sup>3</sup> to 10<sup>4</sup> times higher than the indoor air. This results in a permanent concentration gradient, which is maintained by the long lived decay products of the uranium and thorium series. Radium and radon are the daughter products formed by the radioactive decay of uranium. Each atom of radium decays by ejecting from its nucleus an alpha particle. As the alpha particle is ejected, the newly formed radon atom recoils in the opposite direction. But all the radon atoms may not enter the pore space between the mineral grain of the rock or soil. The chances of the newly formed radon to enter the pore space depend on the proximity of the radium atom to the surface of the mineral grain and also on the direction of the recoil of the radon atom. Under the above mentioned conditions, some fraction of radon atom may reach the pore space of the mineral grain. This fraction is known as emanation fraction (Nazaroff et al., 1988a).

From the site of generation, the radon is readily transported to the air filled pores and then to the soil or building air interface. Radon in the pore space may be transported to the soil air interface by diffusion and advection processes. But in most of the cases the radon is transported by diffusion. From the emanation, followed by the transportation inside the soil, the radon is released to the atmosphere. This release is known as radon exhalation (Ishimori et al., 2013). The emanation and exhalation of radon from the soil or rock depends upon many parameters like radium distribution, grain size, porosity, moisture content etc. of the soil/rock (Nazaroff et al., 1988a; Ishimori

et al., 2013). Because the emanation and exhalation of radon depends mainly on the internal structure of the soil, the applied processing technique of the soil or building material may have significant effect on final exhalation of radon (Sas et al., 2012, 2015; Tuccimei and Moroni, 2006).

The objective of the present study is the determination of terrestrial radionuclide activity, emanation, exhalation activity and to see the effect of the internal structure of soil (pore space and grain size) on radon (<sup>222</sup>Rn) and thoron (<sup>220</sup>Rn) exhalation and emanation rate. We aimed to see the effect of various parameters on the radon emanation, transportation and exhalation. The effect of heat treatment on the exhalation and emanation power of soil has been observed. No such study has been performed in this study area, so the results of this study will serve as a baseline of natural radioactivity of the study area.

#### 2. Study area

The Jaipur and Ajmer districts of Rajasthan are surrounded by many volcanic and igneous rocks. Around these districts, many granitic and igneous granitoid bodies exist. During different natural weathering processes, these elements get transferred to surface and sub surface water levels and contaminate it to different extents.

Geologically, Ajmer district is occupied by Bhilwara and Delhi Supergroup, which are further into several groups and formation. The rocks of Bhilwara Supergroup range extends from Devi in the north-east through Kishangarh to Sarwar area, underlying the marwar plains and comprises metasedimentary sequences with associated magmatic complex and igneous rocks. The main Aravalli range extending from Rupnagar in north to Todgarh in south is occupied by the rocks of Delhi supergroup, which comprises calcareous, argillaceous and arenaceous metasedimentary sequence with associated basic volcanic and igneous rocks. In Jaipur district, Delhi and Bhilwara supergroup are the oldest rock type overlain by quartzite, schists, conglomerates, dolomitic limestone etc. Belonging to Alwar and Ajabgarh group of Delhi supergroup along with granite, pegmatite and amphibolites intrusive of Post Delhi age. Hard rocks in major parts of the district are covered by Quarternary fluvial and Aeolian deposits mainly composed of sand, silt, clay and gravel.

#### 3. Methodology

## 3.1. Sample collection and preparation

3–4 bulk soil samples of 1–1.5 kg weight have been collected from each sampling site. The soil samples have been collected from the undisturbed soil from a depth of 10–15 cm so as to minimize the external influences on results due to human activities. The soil samples have been dried in an oven to a constant mass at a temperature of 105 °C to remove moisture content. The dried samples have been homogenized using pestle and mortar by crushing large soil grains and have been sieved through a mesh of 1 mm to remove the stones and organic material. Then a sieve of mesh size 150 µm has been used to sieve the obtained soil samples. In order to obtain the Radium (<sup>226</sup>Ra) activity in the soil samples, 500 g of each above prepared soil samples have been sealed in the marinelli beakers for about a month to obtain secular equilibrium between <sup>222</sup>Rn and its progenies.

For exhalation and emanation measurement studies, the soil samples have been placed in a cylindrical vessel keeping in mind that the free volume is 10 times higher than the sample volume in order to neglect backdiffusion (Tuccimei and Moroni, 2006; Petropoulos et al., 2001).

#### 3.2. Gamma spectrometry

The <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K activity in the collected soil samples have been determined using NaI(TI) scintillation detector of size 63 mm × 63 mm with multiple channel analyzer. The 661 keV energy photopeak of <sup>137</sup>Cs is used to stabalize the photopeak. The activity

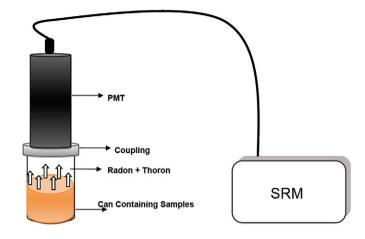


Fig. 1. Experimental setup to measure <sup>222</sup>Rn exhalation and emanation rate.

concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in soil has been determined from the 1764 keV gamma line of <sup>214</sup>Bi, 2610 keV gamma line of <sup>208</sup>Tl and 1460 keV photopeak respectively.

## 3.3. Measurement of $^{222}$ Rn exhalation ( $J_R$ ) and emanation rate ( $E_R$ )

<sup>222</sup>Rn exhalation and emanation measurements have been carried out by coupling the soil sample in a cylindrical chamber to the online radon monitor. The build-up of the radon concentration inside the chamber has been monitored using smart radon monitor (SRM) at different interval of time up to the saturation concentration (Petropoulos et al., 2001; Sahoo et al., 2007).

The radon concentration inside the chamber is given by Eq. (1).

$$C = \frac{J \times M}{V\lambda} \left( 1 - e^{-\lambda t} \right) + Coe^{-\lambda t}$$
<sup>(1)</sup>

where J, M, V,  $\lambda$  and t are the mass exhalation rate, mass of dry sample, effective volume, effective decay constant and measurement time.

Under present investigation conditions, the contribution of leakage rate and back diffusion is negligible because the PMT of the scintillation detector is directly mounted on the cylindrical chamber and it was made air tight using gasket as shown in Fig. 1.

The emanation rate of the <sup>222</sup>Rn has been measured using the growth curve method (Tuccimei and Moroni, 2006; Petropoulos et al., 2001). The sample is connected to active radon monitor and radon concentration is monitored till it reaches the maximum value.

## 3.4. Measurement of $^{220}$ Rn exhalation ( $J_T$ ) and emanation rate ( $E_T$ )

<sup>220</sup>Rn exhalation rate has been determined in the similar manner as done for the <sup>222</sup>Rn. The cylindrical chamber containing the soil samples is connected to the active radon monitor Rad7 in a closed loop and monitored for <sup>220</sup>Rn build up for at least 4 h as shown in Fig. 2.

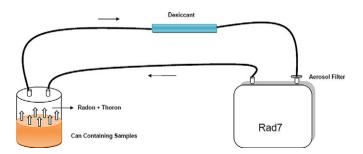


Fig. 2. Experimental setup to measure <sup>220</sup>Rn exhalation and emanation rate.

| Table 1 |  |
|---------|--|
|---------|--|

<sup>226</sup>Ra, <sup>232</sup>Th concentration and <sup>222</sup>Rn, <sup>220</sup>Rn Exhalation and Emanation rate of soil.

| Location      | $C_T (Bq kg^{-1})$ | $C_R$ (Bq kg <sup>-1</sup> ) | $C_{\rm K}$ (Bq kg <sup>-1</sup> ) | $J_T(Bq\;kg^{-1}\;h^{-1})$ | $E_T (kg^{-1} s^{-1})$ | $J_{R}(mBqkg^{-1}h^{-1})$ | $E_R (kg^{-1} s^{-1})$ | Porosity | B.D (g cm <sup>-3</sup> ) | P.D (g cm <sup>-3</sup> ) |
|---------------|--------------------|------------------------------|------------------------------------|----------------------------|------------------------|---------------------------|------------------------|----------|---------------------------|---------------------------|
| Khudiyala     | $130\pm16$         | $73\pm11$                    | $716\pm121$                        | $21.4\pm1.9$               | $99.5\pm8.5$           | $8.31 \pm 0.53$           | $0.19\pm0.01$          | 0.49     | 1.59                      | 3.12                      |
| Mal ki dhani  | $75 \pm 12$        | $61 \pm 10$                  | $622\pm80$                         | $13.3\pm1.3$               | $99.6\pm9.2$           | $7.54\pm0.79$             | $0.12\pm0.01$          | 0.57     | 1.24                      | 2.90                      |
| Peesangan     | $85 \pm 12$        | $69 \pm 11$                  | $753\pm123$                        | $10.0\pm1.4$               | $62.6\pm5.3$           | $9.29\pm0.63$             | $0.19\pm0.01$          | 0.39     | 1.66                      | 2.71                      |
| Chakwada      | $50 \pm 9$         | $52 \pm 9$                   | $875\pm123$                        | $17.9 \pm 1.7$             | $111.2\pm6.4$          | $8.99\pm0.60$             | $0.18\pm0.01$          | 0.52     | 1.31                      | 2.71                      |
| Bhojpura      | $51 \pm 9$         | $51 \pm 9$                   | $905\pm119$                        | $41.2 \pm 2.0$             | $322.9 \pm 21.3$       | $15.86 \pm 0.78$          | $0.34\pm0.02$          | 0.49     | 1.04                      | 2.03                      |
| Harmada       | $76 \pm 12$        | $65 \pm 11$                  | $724\pm123$                        | $13.4 \pm 1.5$             | $88.5 \pm 7.4$         | $10.90\pm0.57$            | $0.25 \pm 0.01$        | 0.57     | 1.18                      | 2.71                      |
| Kanpura       | $71\pm10$          | $63 \pm 10$                  | $887\pm130$                        | $10.0 \pm 1.2$             | $60.6\pm4.5$           | $5.20\pm0.49$             | $0.1\pm0.01$           | 0.67     | 1.35                      | 4.07                      |
| Saipura       | $76 \pm 10$        | $62 \pm 10$                  | $745\pm98$                         | $8.0 \pm 1.1$              | $45.5\pm4.3$           | $7.44\pm0.59$             | $0.16\pm0.01$          | 0.65     | 2.03                      | 5.82                      |
| Jeetawala     | $42 \pm 8$         | $50 \pm 9$                   | $1061\pm125$                       | $18.2\pm1.6$               | $115.5 \pm 7.3$        | $5.36 \pm 0.49$           | $0.11\pm0.01$          | 0.66     | 1.40                      | 4.07                      |
| Bandanwada    | $88 \pm 12$        | $57 \pm 10$                  | $963 \pm 129$                      | $56.8 \pm 2.5$             | $298.3 \pm 20.5$       | $10.67 \pm 0.71$          | $0.21 \pm 0.01$        | 0.42     | 1.62                      | 2.81                      |
| Fatehgarh     | $81 \pm 11$        | $46 \pm 9$                   | $1082\pm130$                       | $54.7~\pm~2.4$             | $341.0\pm22.9$         | $13.04 \pm 0.66$          | $0.26\pm0.01$          | 0.55     | 1.48                      | 3.26                      |
| Borada        | $107\pm14$         | $51 \pm 9$                   | $926\pm126$                        | $21.2 \pm 1.9$             | $132.6 \pm 12.4$       | $10.00\pm0.62$            | $0.21\pm0.01$          | 0.83     | 1.40                      | 8.16                      |
| Shrinagar     | $36 \pm 8$         | $39 \pm 8$                   | $956 \pm 132$                      | $17.1 \pm 1.7$             | $75.9 \pm 5.6$         | $10.47 \pm 0.74$          | $0.27\pm0.02$          | 0.39     | 1.98                      | 3.26                      |
| Chomu         | $40 \pm 7$         | $43 \pm 9$                   | $1016\pm130$                       | $20.7 \pm 1.8$             | $131.2 \pm 12.1$       | $16.31 \pm 0.97$          | $0.40\pm0.02$          | 0.69     | 1.25                      | 4.07                      |
| Achrol        | $30 \pm 7$         | $34 \pm 8$                   | $1000\pm128$                       | $19.4 \pm 1.9$             | $111.1 \pm 11.3$       | $11.87 \pm 0.86$          | $0.24\pm0.02$          | 0.67     | 1.35                      | 4.07                      |
| Daulatpura    | $56 \pm 9$         | $49 \pm 9$                   | $825\pm112$                        | $34.3 \pm 2.1$             | $193.9 \pm 12.8$       | $14.59 \pm 0.94$          | $0.32\pm0.02$          | 0.56     | 1.43                      | 3.26                      |
| Patan         | $145\pm18$         | $70 \pm 11$                  | $877\pm131$                        | 103.8 ± 5.1                | $618.7\pm41.2$         | $16.66 \pm 0.99$          | $0.35\pm0.02$          | 0.57     | 1.35                      | 3.13                      |
| Ajmer         | $48 \pm 9$         | $50 \pm 9$                   | $1012\pm131$                       | $12.0\pm1.4$               | $60.9\pm4.7$           | $16.15 \pm 1.04$          | $0.34\pm0.02$          | 0.60     | 1.62                      | 4.07                      |
| Navalpura     | $39 \pm 8$         | $47 \pm 9$                   | $1007\pm112$                       | $25.5 \pm 2.0$             | $157.2 \pm 14.5$       | $21.04 \pm 1.11$          | $0.5\pm0.03$           | 0.57     | 1.16                      | 2.71                      |
| Ratanpura     | $75 \pm 10$        | $56 \pm 9$                   | $995\pm132$                        | $52.6 \pm 5.6$             | $240.3 \pm 14.4$       | 26.29 ± 1.39              | $0.49\pm0.03$          | 0.60     | 1.62                      | 4.07                      |
| Dudu          | $88 \pm 12$        | $61 \pm 10$                  | $783\pm93$                         | $19.9 \pm 1.8$             | $119.2\pm9.3$          | $10.51 \pm 0.78$          | $0.22\pm0.02$          | 0.57     | 1.25                      | 2.91                      |
| Jaipur        | $36 \pm 4$         | $44 \pm 7$                   | $750\pm100$                        | $27.9 \pm 2.3$             | $139.4 \pm 13.4$       | $16.83 \pm 0.96$          | $0.38\pm0.02$          | 0.68     | 1.34                      | 4.08                      |
| Kotputli      | $40 \pm 9$         | $50 \pm 10$                  | $987\pm120$                        | $23.5 \pm 1.9$             | $117.5 \pm 11.3$       | $7.69 \pm 0.64$           | $0.17\pm0.01$          | 0.65     | 2.03                      | 5.82                      |
| Kekri         | $54 \pm 6$         | $47 \pm 8$                   | $792\pm110$                        | $40.7 \pm 2.2$             | $203.5 \pm 13.7$       | $15.74 \pm 0.95$          | $0.36 \pm 0.02$        | 0.45     | 1.59                      | 2.89                      |
| Bhinai        | $93 \pm 9$         | $56 \pm 8$                   | $854\pm98$                         | $62.6 \pm 3.2$             | $312.8\pm21.4$         | $10.14 \pm 0.75$          | $0.21\pm0.02$          | 0.60     | 1.62                      | 4.06                      |
| Jamwa ramgarh | $55 \pm 11$        | $63 \pm 8$                   | $814\pm121$                        | $12.1 \pm 1.5$             | $60.5\pm8.6$           | $9.63 \pm 0.70$           | $0.21\pm0.02$          | 0.57     | 1.36                      | 3.16                      |
| Nagola        | $64 \pm 8$         | $43 \pm 9$                   | $912\pm113$                        | $67.6 \pm 3.6$             | $338.2 \pm 23.4$       | $12.24 \pm 0.61$          | $0.26 \pm 0.01$        | 0.41     | 1.63                      | 2.76                      |
| Lamba         | $93 \pm 9$         | $68 \pm 8$                   | $925\pm130$                        | $12.7 \pm 1.8$             | $63.4 \pm 6.7$         | $5.43 \pm 0.52$           | $0.12\pm0.01$          | 0.64     | 2.1                       | 5.83                      |
| Bagru         | $48 \pm 8$         | $51 \pm 6$                   | $748\pm96$                         | $25.1 \pm 2.1$             | $125.4 \pm 12.3$       | $8.14 \pm 0.63$           | $0.17 \pm 0.01$        | 0.53     | 1.15                      | 2.45                      |
| Jairam pura   | $50\pm6$           | $47 \pm 7$                   | $912\pm114$                        | $21.9 \pm 1.8$             | $109.6 \pm 12.3$       | $8.62 \pm 0.57$           | $0.19\pm0.01$          | 0.40     | 1.58                      | 2.63                      |
| Magliyawas    | $92 \pm 10$        | $66 \pm 9$                   | $743\pm109$                        | $12.9 \pm 1.5$             | $64.3 \pm 5.6$         | $5.75 \pm 0.52$           | $0.13 \pm 0.01$        | 0.38     | 1.54                      | 2.48                      |
| Dilwara       | $88 \pm 10$        | $73 \pm 9$                   | $812\pm112$                        | $11.2 \pm 1.2$             | $56.2 \pm 4.3$         | $6.01 \pm 0.57$           | $0.13 \pm 0.01$        | 0.43     | 1.63                      | 2.86                      |
| Lamba         | $93 \pm 9$         | $68 \pm 8$                   | $925 \pm 130$                      | $11.9 \pm 1.4$             | $58.4 \pm 3.2$         | $5.62 \pm 0.55$           | $0.12\pm0.01$          | 0.39     | 1.55                      | 2.54                      |
| Shahpura      | 42 ± 8             | $54 \pm 7$                   | 943 ± 110                          | $12.1 \pm 1.3$             | $60.3 \pm 5.3$         | $10.84 \pm 0.81$          | $0.23 \pm 0.02$        | 0.58     | 1.63                      | 3.88                      |

 $^{220}$ Rn emanation rate has been calculated according to the technique developed by Csige et al. (2013). The  $^{220}$ Rn emanation rate is given by Eq. (2).

$$E_{\rm T} = \frac{G}{\lambda x p} \tag{2}$$

where G and  $\rho$  are the  $^{220}\!\text{Rn}$  generation rate and density of sample.

### 3.5. Grain size analysis

Grain size analysis provides us the grain size distribution, percentage of different grain sizes contained in the soil. Distribution of different grain sizes affects the emanation and exhalation property of soil so it is important to carry out the grain size analysis. Sieve analysis has been performed to get the grain size distribution and average grain size of the soil particles. To see the effect of grain size on the Exhalation, Emanation rate of <sup>222</sup>Rn & <sup>220</sup>Rn, the soil has been divided into further categories of grain sizes as x (Grain size: 250  $\mu$ m), y (Grain size: 300–150  $\mu$ m), z (Grain size: 150–75  $\mu$ m), g (Grain size: <75  $\mu$ m).

#### 4. Results and discussion

4.1. Measurement of  $^{226}$ Ra ( $C_R$ ),  $^{232}$ Th ( $C_T$ ) and  $^{40}$ K ( $C_K$ ) concentration in soil

The rocks present in the Earth's crust belong to igneous, sedimentary and metamorphic groups. Activity of the terrestrial radionuclides is highest in shale, deep-sea sediments and phosphate rocks. Except them, igneous rocks are usually more radioactive than the sedimentary ones (Rosler and Lange, 1965). The study area is surrounded by volcanic and igneous rocks which make it important for the radioactivity measurements. The level of natural radionuclides in the soil is very much dependent on the parent rock from which it is formed. Due to the weathering of the  $^{226}$ Ra from the host rocks, it can be transported and deposited as loess, silt placers and tertiary soil. (Iyengar, 1990). Because of the extreme insolubility of  $^{232}$ Th in water, it cannot be transported by water during weathering process. So it remains concentrated in its parent rock and from here it is transported in the form of colloidal suspension. Due to physical bond to the surface of colloids, it is preferentially found in gravel deposits and coarse sandstones (Michel, 1990). The radium content of the soil is given as its activity per unit dry mass (Bq kg<sup>-1</sup>). The calculated  $^{226}$ Ra and  $^{232}$ Th radionulide concentrations for the 34 samples collected from the study area are reported in Table 1. It can be clearly seen from the observed data that the  $^{232}$ Th activity in soil is higher than that of  $^{226}$ Ra and ranged between 30 Bq kg<sup>-1</sup>

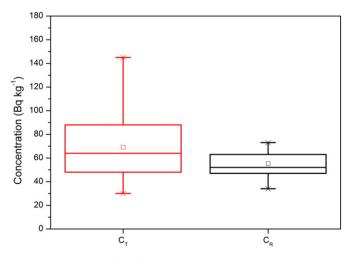


Fig. 3. <sup>226</sup>Ra, <sup>232</sup>Th concentration variation in soil.

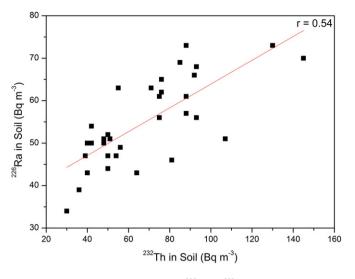


Fig. 4. Correlation between <sup>226</sup>Ra and <sup>232</sup>Th in soil.

and 145 Bq kg<sup>-1</sup> with an average of 69 Bq kg<sup>-1</sup>. On the other hand, the activity concentration of <sup>226</sup>Ra is lower than that of <sup>232</sup>Th and shows the variation between 34 Bq kg<sup>-1</sup> and 73 Bq kg<sup>-1</sup> with an average of 55 Bq kg<sup>-1</sup> (Fig. 3). The activity concentration of <sup>40</sup>K in the studied soil samples varies from 622 Bq kg<sup>-1</sup> to 1082 Bq kg<sup>-1</sup>, with an average value of 874 Bq kg<sup>-1</sup>.

In order to find the extent of togetherness of these radionuclides at a particular location, a correlation study has been carried between these radionuclides. The regression carried was between <sup>226</sup>Ra and <sup>232</sup>Th and the scatter plot for this pair is shown in Fig. 4. These type of positive correlations have been reported in the literature between <sup>232</sup>Th and <sup>238</sup>U: <sup>226</sup>Ra and <sup>40</sup>K; <sup>232</sup>Th and <sup>40</sup>K. (Kannan et al., 2002.; Quindos et al., 1994) but some studies has also shown the negative correlations between <sup>232</sup>Th and <sup>40</sup>K (Mehra et al., 2010). This may be due to the different farm practices involving in improving the soil fertility by use of fly ash in appropriate combination with organic matter and chemical fertilizer (Mittra et al., 2003) which may result in the altering of the distribution and concentration of these radionuclides from their natural order in the soil of the particular area. In our case, the observed moderate linear positive correlation coefficient between <sup>226</sup>Ra and <sup>232</sup>Th activity concentrations (Fig. 4) indicate a statistically significant and moderately strong relationship. Similar types of results have been reported by Kovács et al., 2013.

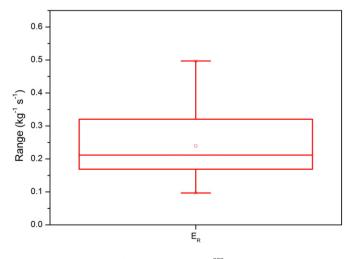


Fig. 5. Emanation rate of <sup>222</sup>Rn.

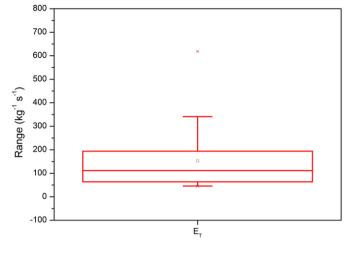


Fig. 6. Emanation rate of <sup>220</sup>Rn.

4.2. <sup>222</sup>Rn, <sup>220</sup>Rn exhalation and emanation rate

The formation, transportation and emission of the radon (<sup>222</sup>Rn and <sup>220</sup>Rn) from soil to air depend upon many influencing factors. These factors are the concentration of the parent radionuclide isotopes in the mineral grains, probability of the generated <sup>222</sup>Rn and <sup>220</sup>Rn from the decay of parent isotope to reach the pore space of the mineral grain (emanation) and the diffusive or advective flow of the emanated <sup>222</sup>Rn and <sup>220</sup>Rn in the interstitial spaces between the grains. In order to enter the atmosphere, some fraction of the generated and transported radon atoms within the soil reaches the soil air interface, which is known as the exhaled fraction of <sup>222</sup>Rn or <sup>220</sup>Rn (Moed et al., 1988).

With the use of the closed chamber technique, the growth of  $^{222}$ Rn in the sealed container has been monitored for the measurements of the  $^{222}$ Rn emanation and exhalation rate using Eq. (1). However the  $^{220}$ Rn emanation rate has been calculated through the thoron generation rates measurements using Eq. (2) by the same closed chamber technique. The results of the various  $^{222}$ Rn,  $^{220}$ Rn emanation and exhalation rates in various soil samples of the study area are presented in Table 1 and their statistical variation is shown in Figs. 5, 6 and 7. The ranges of the  $^{222}$ Rn emanation and exhalation rate per unit mass has comes out to be 0.10–0.50 kg<sup>-1</sup> s<sup>-1</sup> and 5.20–26.29 mBq kg<sup>-1</sup> h<sup>-1</sup> respectively. The individual  $^{220}$ Rn emanation and exhalation data should be handled carefully. The calculated values of the  $^{220}$ Rn emanation

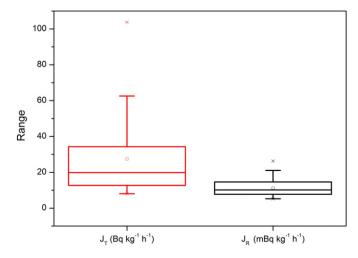


Fig. 7. Exhalation rate of <sup>222</sup>Rn, <sup>220</sup>Rn.

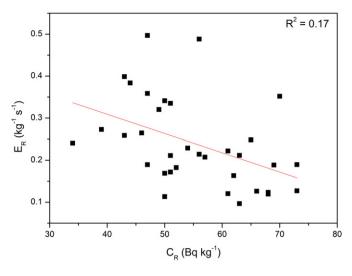
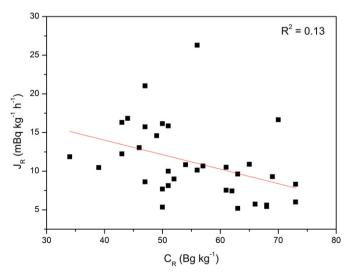


Fig. 8. Correlation between E<sub>R</sub> and C<sub>R</sub>.

and exhalation rates show the variation from 45.51 to 618.74 kg<sup>-1</sup> s<sup>-1</sup> and from 8.01 to 103.77 Bq h<sup>-1</sup> kg<sup>-1</sup> respectively.

No significant relationship has been observed between the <sup>222</sup>Rn emanation, exhalation rate with the <sup>226</sup>Ra concentration (Figs. 8, 9). Similar type of results has been observed for the <sup>220</sup>Rn emanation, exhalation rate with the concentration of <sup>232</sup>Th in the studied soil samples of the study area (Figs. 10, 11). In case of the <sup>220</sup>Rn, the emanated and exhaled fraction is dependent on many soil parameters like temperature, moisture etc. This type of <sup>220</sup>Rn behaviour is also obvious due to its much larger decay constant as compared to <sup>222</sup>Rn. Non-significant correlation coefficient between <sup>222</sup>Rn and <sup>226</sup>Ra has also been reported in other studies (Baykara et al., 2005; Imme et al., 2014). The transportation of <sup>222</sup>Rn from its <sup>226</sup>Ra source in the mineral grains of soil is carried by its emanation which depends upon <sup>226</sup>Ra distribution in the grains, water content of the grains and grin size followed by transportation in the pore space and then exhalation which also depends upon water content, porosity, grain size and soil temperature (Hassan et al., 2009; Baixeras et al., 2001; Ramola and Choubey, 2003; Righi and Bruzzi, 2006). The distribution of the grain size of the soil also influences its permeability, which will in turn affect its <sup>222</sup>Rn and <sup>220</sup>Rn exhalation. Due to the dependence of the so many parameters on the <sup>222</sup>Rn, <sup>220</sup>Rn emanation and exhalation rate, their correlation with parent radionuclide concentration is only possible in case of the controlled experimental conditions.



**Fig. 9.** Correlation between J<sub>R</sub> and C<sub>R</sub>.

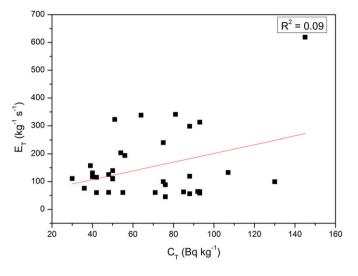


Fig. 10. Correlation between  $E_T$  and  $C_T$ .

#### 4.3. Effect of soil parameters on emanation of <sup>222</sup>Rn and <sup>220</sup>Rn

Emanation of <sup>222</sup>Rn and <sup>220</sup>Rn from soil grains is affected by many soil properties and parameters. Radium distribution in the soil grains, grain size, and porosity are some important parameters to study the effects of these on emanation. The experimental results of the dependence of emanation rate on these parameters have been discussed below.

#### 4.3.1. Radium distribution

The results of <sup>226</sup>Ra concentration versus grain size of the soil sample are presented and plotted in Fig. 12. These results show the distribution of the <sup>226</sup>Ra over the surface layer rather than over the grain volume. Each soil sample has been divided into different grain size fraction, prior to the measurement. The <sup>226</sup>Ra concentration shows an increasing trend with the decreasing grain size of soil sample approximately upto 150 µm. For the larger grains, due to relatively smaller surface area per grain, the amount of the <sup>226</sup>Ra nuclides adsorbed on the grain surface is negligible as compared to the amount of <sup>226</sup>Ra present originally in soil. In case of fine soil grains, the surface area to volume ratio becomes larger and the <sup>226</sup>Ra content become more concentrated at the surface layer. So the smaller grains contain more <sup>226</sup>Ra than the larger grains. The obtained experimental results of the <sup>226</sup>Ra content of soil as the

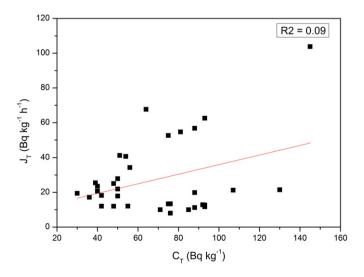


Fig. 11. Correlation between J<sub>T</sub> and C<sub>T</sub>.

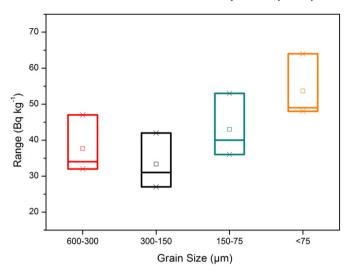


Fig. 12. Variation of radium content with grain size.

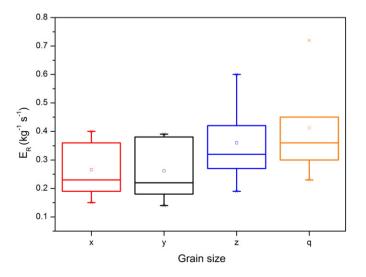
function of grain size are in agreement with the results of the other studies reported in literature (Breitner et al., 2010; Blanco Rodriguez et al., 2008).

#### 4.3.2. Grain size

<sup>222</sup>Rn, <sup>220</sup>Rn emanation rate measurements have been carried out for the soil divided into different grain size composition. The divided individual soil sample has the grain size of 250 μm, grain size lying between 300 and 150 μm, 150–75 μm and <75 μm. Some other authors have also shown the similar variation of the emanation with the grain size (Markkanen and Arvela, 1992; Hosoda et al., 2008; Breitner et al., 2010). The results of the variation of the emanation rate of <sup>222</sup>Rn with grain size shows the decreasing trend upto the grain size of 150 μm then it becomes constant. In case of emanation rate of <sup>220</sup>Rn a fall has been observed when the grain size exceeds the diameter of 150 μm. The values of  $E_T$  below and above these diameters are approximately constant (Figs. 13, 14).

#### 4.4. Effect of soil parameters on exhalation rate

Like emanation, exhalation of <sup>222</sup>Rn and <sup>220</sup>Rn are also a function of various soil parameters discussed below:



**Fig. 13.** Variation of  $E_R$  with grain size.

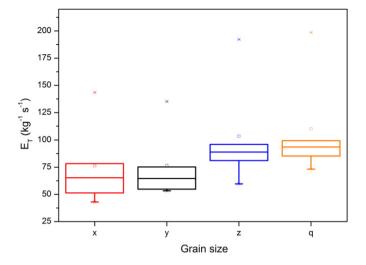


Fig. 14. Variation of E<sub>T</sub> with grain size.

#### 4.4.1. The porosity

The experimental results of the variation of radon exhalation rate with porosity has shown steadily increasing trend with increase in the porosity of the same medium. In case of low porosity, soil grains are closed to each other. So, the radon atoms in the pore space are unable to find their path to the outer environment. With the increase in porosity, the pore volume of the soil increases hence the exhalation rate also increases (Figs. 15, 16).

#### 4.4.2. The grain size

The results show as the grain size of the soil sample increases, the exhalation rate of both <sup>222</sup>Rn and <sup>220</sup>Rn decreases (Figs. 17, 18). The dependency of the emanation rate of radon on grain size as explained in the figs. 12, 13 and the decrease in the mass exhalation rate with decrease in porosity of the soil as shown in Figs. 15 and 16 may explain the possible reasons of dependence of exhalation rate of 222Rn and <sup>220</sup>Rn on grain size. Hassan et al., 2009 and cited references have also observed the similar type of control of grain size over the exhalation rate of radon.

#### 5. Conclusions

The Soil samples collected from the different locations of the study area have been analysed for terrestrial radionuclide content (<sup>232</sup>Th,

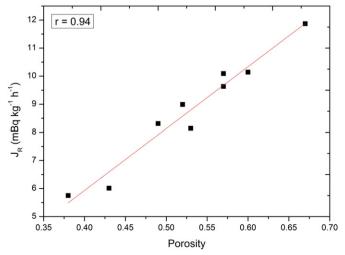


Fig. 15. Variation of J<sub>R</sub> with porosity.

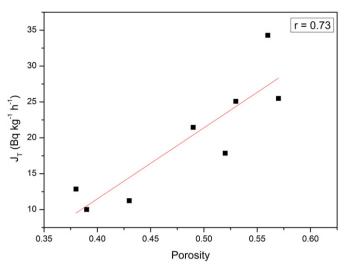
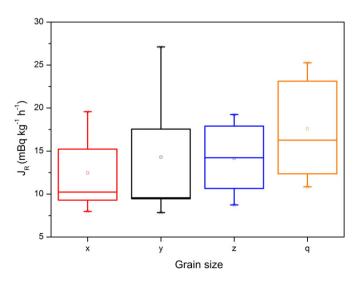


Fig. 16. Variation of J<sub>T</sub> with porosity.

<sup>226</sup>Ra, <sup>40</sup>K); Exhalation, Emanation rate and their correlation with different soil parameters.

The average <sup>226</sup>Ra and <sup>232</sup>Th activity of the investigated soil samples are 55 Bq kg<sup>-1</sup> and 69 Bq kg<sup>-1</sup> respectively, which is more than the world average of <sup>226</sup>Ra (32 Bq kg<sup>-1</sup>) and <sup>232</sup>Th in soil (45 Bq kg<sup>-1</sup>) (UNSCEAR, 2010). The high values of the <sup>226</sup>Ra and <sup>232</sup>Th radionuclide's in the soil samples is associated with the presence of the granitic and gratinoid bodies in the study area. The activity concentration of these radionuclides in the soil samples of Jaipur district is relatively low due to the nonpresence of the parent rock of the igneous or volcanic origin. Investigations of the variables known to impact <sup>222</sup>Rn exhalation have been performed and are presented in the present study. The results have shown the effects of soil parameters on Exhalation and Emanation rate of radon from soil. From the studies reported in literature, it has been shown that the moisture is one of the most dominating variables that impact the <sup>222</sup>Rn exhalation and emanation rates (Breitner et al., 2010; Hosoda et al., 2010; Janik et al., 2015). But it is cleared from the experimental results of this study that apart from the <sup>226</sup>Ra content of the soil samples, the <sup>222</sup>Rn, <sup>220</sup>Rn exhalation and emanation rates can be greatly affected by the soil parameters such as porosity and grain size. The results of the present investigation support the theory that the <sup>222</sup>Rn exhalation and emanations are not dependent on the <sup>226</sup>Ra content of the material only.



**Fig. 17.** Variation of  $J_R$  with grain size.

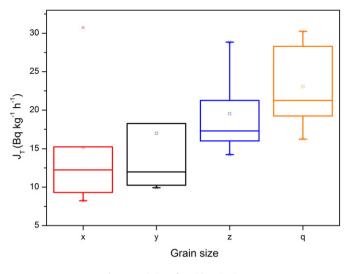


Fig. 18. Variation of  $J_T$  with grain size.

#### **Conflict of interest**

Authors declare no conflict of interest.

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