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Variability of radon and thoron equilibrium factors in indoor environment of Garhwal Himalaya

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ABSTRACT

The measurements of radon, thoron and their progeny concentrations have been carried out in the dwellings of Uttarkashi and Tehri districts of Garhwal Himalaya, India using LR-115 detector based pinhole dosimeter and DRPS/DTPS techniques. The equilibrium factors for radon, thoron and their progeny were calculated by using the values measured with these techniques. The average values of equilibrium factor between radon and its progeny have been found to be 0.44, 0.39, 0.39 and 0.28 for rainy, autumn, winter and summer seasons, respectively. For thoron and its progeny, the average values of equilibrium factor have been found to be 0.04, 0.04, 0.04 and 0.03 for rainy, autumn, winter and summer seasons, respectively. The equilibrium factor between radon and its progeny has been found to be dependent on the seasonal changes. However, the equilibrium factor for thoron and progeny has been found to be same for rainy, autumn and winter seasons but slightly different for summer season. The annual average equilibrium factors for radon and thoron have been found to vary from 0.23 to 0.80 with an average of 0.42 and from 0.01 to 0.29 with an average of 0.07, respectively. The detailed discussion of the measurement techniques and the explanation for the results obtained is given in the paper.

1. Introduction

Radon, thoron and their decay products are present in the indoor environment since their parent nuclei radium and thorium are present in building materials and the soil. It is well known that the inhalation of radon, thoron and their decay products contributes a major part (more than 50%) of the natural background radiation dose to the humans (UNSCEAR, 2008). Further, in the indoor environment, the inhalation doses due to the radon and thoron are predominantly contributed from their decay product concentrations in the indoor environment. The estimation of equilibrium factors for radon, thoron and their progeny is very important for assessing the radiation dose received from the inhalation of radon, thoron and their progeny. Therefore, it is very essential to carry out the systematic long terms measurements of the equilibrium factors for radon and its progeny and thoron and its progeny in the dwellings of the general public.

In case of radon exposure, the short lived radon progeny imparts radiation dose to lungs mainly and not the gas concentration itself. Radiation exposure due to radon progeny is estimated as the product of potential alpha energy concentration (PAEC) and the exposure time. The ratio of potential alpha energy concentration to the radon concentration can be substituted by the equilibrium factor (F), which is expressed as (Leung et al., 2006):

$$F = \{0.105\,C_1 + 0.515\,C_2 + 0.380\,C_3\}/C_o$$

where, C_o, C_1, C_2 and C_3 indicate the activity concentrations (in Bq/m^3) of ^{222}Rn, ^{218}Po, ^{214}Pb and ^{214}Bi, respectively.

In the past, radiation dose to the lungs due to exposure of radon progeny has been estimated by first measuring the radon gas concentration and then applying the equilibrium factor, considering the assumed value (0.4) of equilibrium factor for radon and its progeny (ICRP, 1991; UNSCEAR, 2008). However, the radon progeny and hence the equilibrium factor depends largely on the environmental conditions such as hours and modes of ventilation, humidity, etc (Porstendorfer, 1984; Jilek et al., 2010). The equilibrium factor has also been found to vary with time and place (Nikezic and Yu, 2005; Ramola et al., 2003; Yu and Nikezic, 2011; Yu et al., 1996). The ventilation conditions of a building and the

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plating out of radon and thoron progeny atoms onto surfaces also affect the equilibrium factors for radon and thoron (Ramola et al., 2003). In contrast, thoron equilibrium factor varies significantly even for the same environment. This is mainly due to wide variation of thoron concentration arising from its short lived nature. The very short half life of thoron results in non-uniformity of thoron concentration in the indoor environment. Hence it is not advisable to estimate EEC of thoron using the gas concentration and equilibrium factor. Therefore assumed worldwide value of equilibrium factors cannot reflect the actual results and there is a need of direct measurement of radon and thoron decay products concentrations to assess the actual dose received by the general public due to the exposure of progeny. In this study, direct measurements of the decay products and gas concentrations were carried out by using direct progeny sensors and pin-hole dosimeter technique, respectively. The equilibrium factor was then calculated simply by dividing progeny concentration by the gas concentration. The measurements have been taken in 87 houses of Uttarkashi and Tehri areas of Garhwal Himalaya for first quarter (rainy season) and then 57 houses out of these 87 were chosen for seasonal variations.

2. Study area

The Geographical maps of the study area are shown in the Figs. 1 and 2. The study area comprises of Tehri Garhwal and Uttarkashi districts of Uttarakhand, India. The map was prepared with Surfer software using Lambert Conformal Conical (LCC) according to NNRMS (2005) and transform formulae (Snyder, 1987). The sampling locations are shown in Fig. 2.

3. Experimental methods

3.1. Measurement of indoor ^{222}Rn and ^{220}Rn concentrations

Measurements of ^{222}Rn and ^{220}Rn were carried out by LR-115 Type II detector based pin-hole dosimeter technique. The dosimeter is a cylindrical plastic chamber and consists of two equal compartments separated by a central disc, each compartment having a length of 4.1 cm and radius 3.1 cm. Four pin-holes, each having a length of 2 mm and 1 mm diameter are made on this circular disk in order to discriminate ^{220}Rn. The dosimeter has only one entrance through which the gas enters the first chamber namely “radon + thoron” compartment through a 0.56 μm glass fibre filter paper and subsequently diffuses to second chamber called “radon” chamber cutting off the entry of thoron into this chamber because of its very short half-life of 55.6 s compared to that of radon (3.825 days). The LR-115 detector films are fixed at the end of each compartment. The device has been calibrated in a laboratory facility at Bhabha Atomic Research Centre, Mumbai in order to find a correlation between tracks registered on the detector films and the concentration of radon/thoron (Sahoo et al., 2013). The alpha emissions from radon and thoron produce the tracks on LR-115 detector film placed at the end of first chamber while as there is only radon and not thoron in the second chamber, the tracks are registered on the LR-115 detector film placed at the top of this chamber due to the alpha emissions of radon only. The dosimeters were suspended indoor overhead on the ceiling at the minimum height of 1.5 m from the ground and at least 10 cm away from any wall surface for a period of about 3 months.

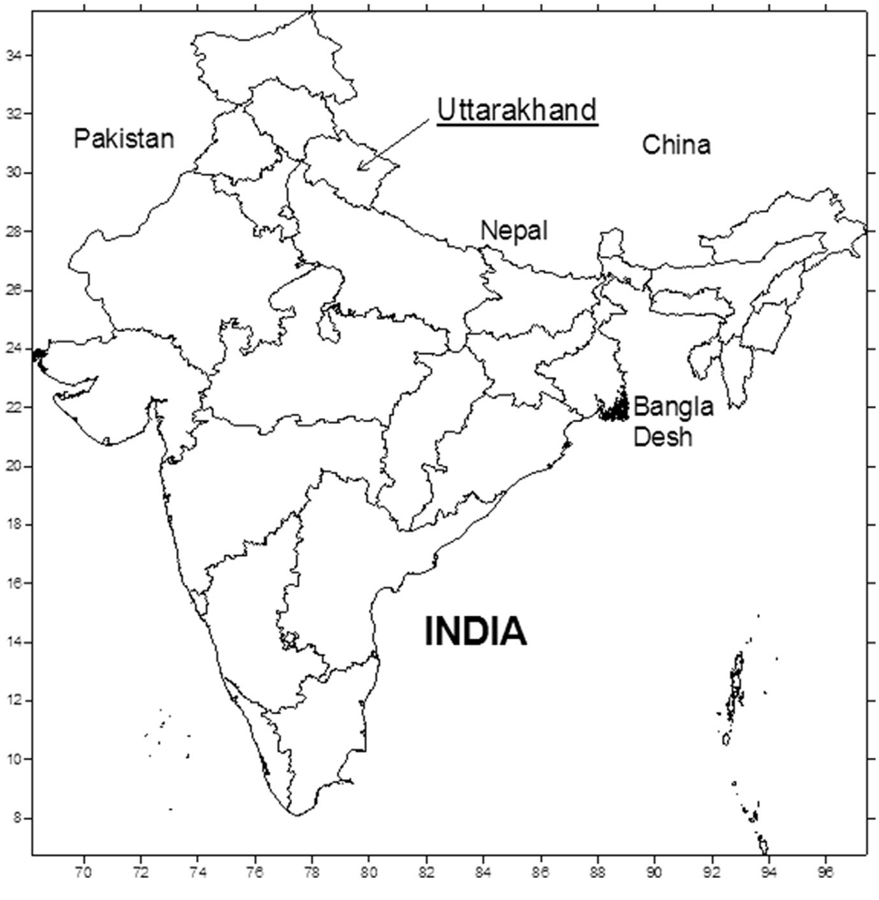


Fig. 1. Shape of India in geographical coordinates.

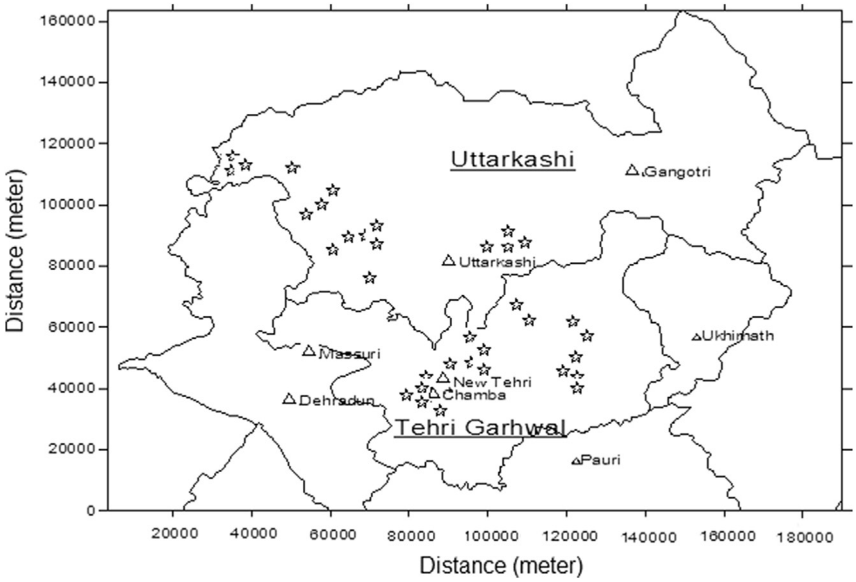


Fig. 2. Map of Uttarakhand showing the sampling sites covering Tehri and Uttarkashi districts.

3.2. Measurement of ^{222}Rn and ^{220}Rn progeny

Cellulose nitrate (LR-115 Type II) solid state nuclear track detector deposition based direct progeny sensor technique has been used for the measurement of ^{222}Rn and ^{220}Rn progeny. Direct progeny sensors are made of passive detectors (LR-115 type II) mounted with absorbers of appropriate thickness. For thoron (^{220}Rn) progeny, the absorber is aluminium Mylar of 50 μm thickness, which selectively records the tracks due to alpha particles emitted from ^{212}Po (α energy 8.78 MeV). For radon (^{222}Rn) progeny, the absorber is a combination of aluminized Mylar (25 μm) and cellulose nitrate film (12 μm) of effective thickness 37 μm, which mainly detects alpha particles emitted from ^{214}Po (α energy 7.69 MeV) and ^{212}Po (α energy 8.78 MeV). The sensor, which is used for the detection of thoron (^{220}Rn) progeny, is known as Direct Thoron Progeny Sensor (DTPS) and the sensor which used for the detection of radon (^{222}Rn) progeny is known as Direct Radon Progeny Sensor (DRPS). The basic principle of the operation of these sensors is that the LR-115 detector detects the alpha particles emitted from the deposited progeny atoms. In calculating the progeny concentrations, the track density obtained using DTPS can be used directly to calculate the equilibrium equivalent thoron concentration (EETC), since the comparatively larger thickness (50 μm) of the absorber used in DTPS does not allow radon progeny to pass through it and hence there is no interference of radon progeny to thoron progeny. In case of equilibrium equivalent radon concentration (EERC), since α energy of ^{212}Po (thoron progeny) is higher than that of ^{214}Po (radon progeny), the alpha particles emitted from both radon progeny as well as from thoron progeny pass through the absorber (37 μm) used in the DRPS and hence to calculate EERC, the tracks produced by the thoron progeny are eliminated as calculated from DTPS, using the equation (Mishra et al., 2009):

$$\text{Tracks}_{\text{DRPS}}^{\text{Only Rn Progeny}} = \text{Tracks}_{\text{DRPS}}^{\text{Total}} - \frac{\eta_{\text{RT}}}{\eta_{\text{TT}}}\text{Tracks}_{\text{DTPS}}^{\text{Total}}$$

where, η_{RT} and η_{TT} are the track registration efficiencies of thoron progeny in DRPS and that in DTPS, respectively, $\text{Tracks}_{\text{DRPS}}^{\text{Only Rn Progeny}}$ is the tracks density recorded on DRPS due to only radon progeny and $\text{Tracks}_{\text{DRPS}}^{\text{Total}}$ and $\text{Tracks}_{\text{DTPS}}^{\text{Total}}$ are the abbreviations for total track density recorded on DRPS and DTPS, respectively.

Formulae used to calculate EETC and EERC (Mishra and Mayya, 2008; Mishra et al., 2009) are given as follows:

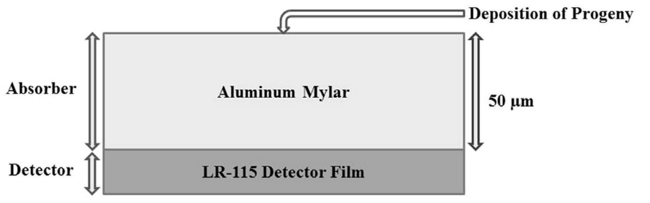
$$\text{EETC}\left(\frac{\text{Bq}}{\text{m}^3}\right) = \frac{\text{Tracks}_{\text{DTPS}}^{\text{Total}}}{\text{k}_\text{T} \times \text{Exposure period(days)}}, \text{ and}$$

$$\text{EERC}\left(\frac{\text{Bq}}{\text{m}^3}\right) = \frac{\text{Tracks}_{\text{DRPS}}^{\text{Only Rn Progeny}}}{\text{k}_\text{R} \times \text{Exposure period(days)}}$$

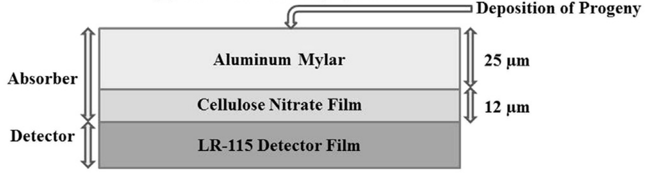
where, k_T and k_R are calibration factors (sensitivity factors) for DTPS and DRPS, respectively. The values of sensitivity factors for DTPS and DRPS in natural environment have been calculated by Mishra et al. (2010) to be equal to 0.94 Tracks $\text{cm}^{-2}\text{d}^{-1}$/EETC (Bq m^{-3}) for DTPS and 0.09 Tracks cm^{-2}/EERC (Bq m^{-3}) for DRPS. The schematic diagram of the direct progeny sensing system is given in the Fig. 3.

3.3. Chemical processing and analysis of LR-115

The exposed LR-115 detector films were etched in an etching



(A) Direct Thoron Progeny Sensor



(B) Direct Radon Progeny Sensor

Fig. 3. Schematic diagram of direct progeny sensors.

bath using 2.5 N NaOH solution at 60 °C temperature for 90 min without stirring. The tracks recorded on the films were then counted using spark counter, which is an electronic counter operating at high voltage. The resulting average track densities were converted into radon, thoron and progeny concentrations using calibration factors discussed above. The equilibrium factor for radon and its progeny and thoron and its progeny were then simply calculated by using the following expressions:

$$\text{Equilibrium Factor for Radon } (F_{Rn}) = \frac{\text{EERC}}{\text{Radon Concentration}}, \text{ and}$$

$$\text{Equilibrium Factor for Thoron } (F_{Tn}) = \frac{\text{EETC}}{\text{Thoron Concentration}}$$

4. Results and discussions

The experimentally determined values of radon concentration, thoron concentration, EERC, EETC, equilibrium factor for radon and progeny (F_{Rn}) and equilibrium factor for thoron and progeny (F_{Tn}) in the 87 dwellings for one quarter (rainy season) are given in the Table 1. The indoor radon concentration and equilibrium equivalent radon concentration (EERC) in the dwellings of the study area have been found to vary from 13 ± 3 Bq/m^3 to 291 ± 13 Bq/m^3 with an average value of 99 ± 59 Bq/m^3 and 3.2 ± 0.6 Bq/m^3 to 191 ± 4.8 Bq/m^3 with an average value of 52 ± 36 Bq/m^3, respectively. The indoor thoron concentration and equilibrium equivalent thoron concentration (EETC) have been found to vary from 0.8 ± 0.6 Bq/m^3 to 141 ± 10 Bq/m^3 with an average value of 42 ± 34 Bq/m^3 and 0.1 ± 0.03 Bq/m^3 to 9.6 ± 0.4 Bq/m^3 with an average value of 2.2 ± 1.7 Bq/m^3, respectively. The equilibrium factor for radon and its progeny has been found to vary from 0.10 to 0.90 with its average value of 0.44 and for thoron and its progeny this factor varies from 0.01 to 0.63 with its average value of 0.05. The equilibrium factor for radon and its progeny was found to be $\leq$0.30 in 17 houses and >0.6 in 25 houses (Fig. 4). However, in remaining 45 houses, this factor lies in the range of 0.30–0.60. The equilibrium factor for thoron and its progeny has been found in the range of 0.10–0.30 in 11 houses and >0.30 in 7 houses. In remaining 69 houses, the equilibrium factor was found to be $\leq$0.10 (Fig. 5).

Out of 87 dwellings sampled for the first quarter (rainy season) of the year, measurements were repeated in 57 dwellings for next three seasons of the year. The seasonal variations of radon, thoron, EERC, EETC, F_{Rn} and F_{Tn} are shown in the Table 2. The average value of radon concentration has been found to be maximum in winter season and minimum in summer season while for thoron concentration, it has been found to be maximum in autumn and minimum in summer season. The comparatively higher average value of radon concentration in the winter season as compared to other seasons is due to the poor ventilation between indoor and outdoor environments of the study area in this season (Ramola et al., 1998). The people used to keep the doors and windows of their houses closed during winter season to conserve the heat inside, which results in poor ventilation between indoor and outdoor environments. On the other hand, the comparatively lower average value of radon concentration in the summer season is due to the poor ventilation between indoor and outdoor environments. Due to the higher average temperature during summer season, there is higher air exchange rate between the indoor and outdoor environments, which results in good ventilation between indoor and outdoor environments. However, the ventilation condition was not found to influence the indoor thoron concentration significantly because of its very short half-life, resulting in an inhomogeneous spatial distribution of thoron in the room.

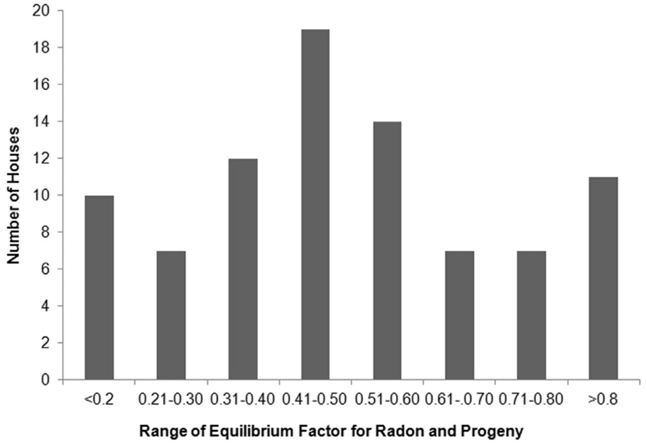


Fig. 4. Frequency distribution of equilibrium factor between radon and progeny in the houses of study area for rainy season.

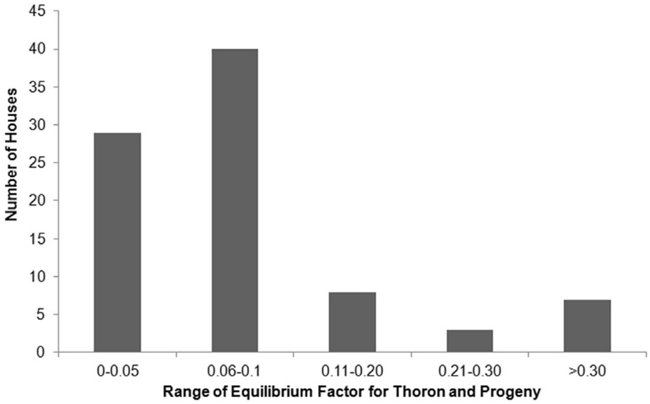


Fig. 5. Frequency distribution of equilibrium factor between thoron and progeny in the houses of study area for rainy season.

Table 1
Experimentally determined values of radon concentration, thoron concentration, EERC, EETC and equilibrium factors for radon and thoron in 87 dwellings of the study area for first quarter (rainy season) of the year.

Experimentally measured values of radon/thoron concentrations, EERC, EETC and equilibrium factors			
	Minimum	Maximum	Average ± SD
Rn-222 Concentration (Bq/m^3)	13 ± 3	291 ± 13	99 ± 59
Rn-220 Concentration (Bq/m^3)	0.8 ± 0.6	141 ± 10	42 ± 34
EERC (Bq/m^3)	3.2 ± 0.6	191 ± 4.8	52 ± 36
EETC (Bq/m^3)	0.1 ± 0.03	9.6 ± 0.4	2.2 ± 1.7
"F_{Rn}" factor for^{222}Rn	0.10	0.90	0.44
"F_{Tn}" factor for^{220}Rn	0.01	0.63	0.05

The average values of EERC and EETC have been found maximum in autumn season and minimum in summer season. The average value of equilibrium factor for radon and its progeny has been found maximum in the rainy and minimum in the summer. However, the equilibrium factor for thoron and its progeny has been found to be same for rainy, autumn and winter seasons but slightly different for summer season.

The annual average equilibrium factors for radon and thoron have been found to vary from 0.23 to 0.80 with an average of 0.42. While for thoron and progeny it varies from 0.01 to 0.29 with an average of 0.07. Among 57 dwellings studied for seasonal variations, the annual average value of equilibrium factor for radon and its progeny was found to be $\leq$0.30 in 9 houses and >0.60 in 7

Table 2
Seasonal variations of radon concentration, thoron concentration, EERC, EETC, and equilibrium factors for radon and thoron in 57 dwellings of the study area.

	Rainy			Autumn			Winter			Summer			Annual average		
	Min	Max	AM ± SD	Min	Max	AM ± SD	Min	Max	AM ± SD	Min	Max	AM ± SD	Min	Max	AM
^{222}Rn Concentration (Bq/m^3)	25 ± 4	259 ± 12	111 ± 57	23 ± 4	668 ± 20	145 ± 130	31 ± 4.3	821 ± 22	146 ± 127	14 ± 3	252 ± 12	88 ± 50	27	397	122
^{220}Rn Concentration (Bq/m^3)	0.8 ± 0.7	547 ± 19	64 ± 104	2 ± 1	584 ± 20	106 ± 140	3 ± 1	367 ± 16	74 ± 90	2 ± 1	212 ± 12	42 ± 36	10	213	69
EERC (Bq/m^3)	3.2 ± 0.6	145.5 ± 4.2	56.5 ± 34.2	12.5 ± 1.2	374.4 ± 6.8	71 ± 75	9.9 ± 2	189 ± 4.8	67 ± 46	4.1 ± 0.7	153.2 ± 4.4	31 ± 32	16	154	56
EETC (Bq/m^3)	0.06 ± 0.03	11.3 ± 0.37	2.4 ± 2.2	0.32 ± 0.06	14.2 ± 0.4	3.9 ± 4	0.1 ± 0.03	9.6 ± 0.3	2.6 ± 1.9	0.1 ± 0.04	6.2 ± 0.27	1.3 ± 1.2	0.2	8	2.5
"F_{Rn}" factor for ^{222}Rn	0.10	0.90	0.44	0.16	0.92	0.39	0.12	0.85	0.39	0.10	0.83	0.28	0.23	0.80	0.42
"F_{Tn}" factor for ^{220}Rn	0.01	0.63	0.04	0.01	0.53	0.04	0.01	0.80	0.04	0.01	0.27	0.03	0.01	0.29	0.07

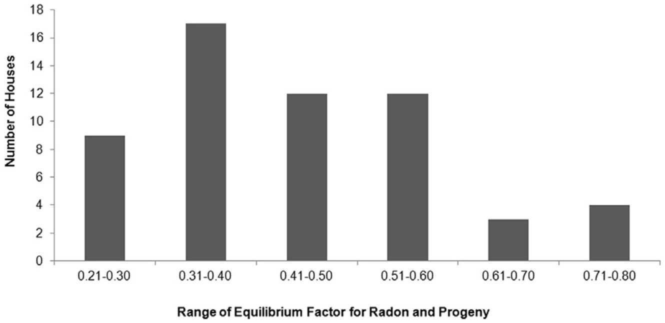


Fig. 6. Frequency distribution of annual average equilibrium factor between radon and progeny in the houses of study area.

houses (Fig. 6). The annual average value of equilibrium factor in the remaining 41 houses has been found in the range 0.30–0.60. The annual average value of equilibrium factor for thoron and its progeny was found to be in the range 0.11–0.20 in the 14 houses and >0.20 in 3 houses (Fig. 7). The annual average value of equilibrium factor in the remaining 40 houses has been found to be in the range of 0.01–0.10.

The experimentally determined annual average value of the equilibrium factor for radon and its progeny (0.42) has found slightly above the globally assumed value of equilibrium factor (0.4) as reported in UNSCEAR (2008). However, this value with improved methodology is found higher than the previously reported value (0.28) for Garhwal Himalaya (Ramola et al., 2003). The annual average value of equilibrium factor for thoron and its progeny (0.07) has been found comparable with world's average value (0.1) as reported in UNSCEAR (1993) and previously determined value (0.09) for Garhwal Himalaya (Ramola et al., 2003). These results show that the values of radon and thoron equilibrium factors are reproducible in the study area with new pin-hole radon dosimeter and DTPS/DRPS.

5. Conclusion

The equilibrium factor for radon and its progeny has been found to be dependent on the seasons. However, in case of thoron and its progeny, equilibrium factor has been found to be same for rainy, autumn and winter seasons with a slight difference for summer season. The annual average equilibrium factors for radon and its progeny and thoron and its progeny were found comparable with the world's average values. However, the value of

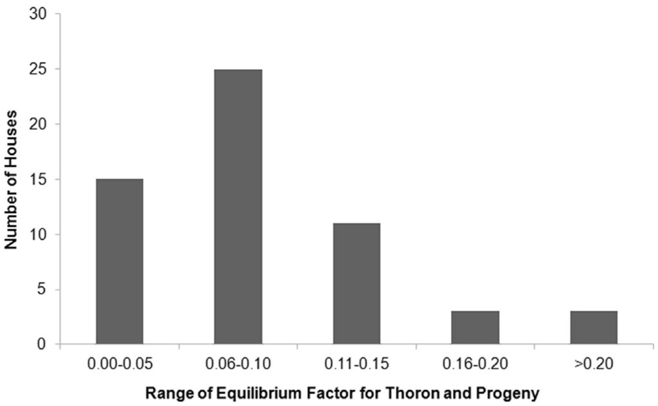


Fig. 7. Frequency distribution of annual average equilibrium factor between thoron and progeny in the houses of study area.

equilibrium factor for radon and its progeny is recorded higher than the previously determined value (0.28) for the study area. The deviation from previously reported value of equilibrium factor may be due to use of improved methodology. Moreover, the selection of houses may also have produced the different results. It was found that the radon progeny and the equilibrium factor depend largely on the environmental conditions, which may results in the variation in dose calculation. The large variation in measured values of equilibrium factor suggests that while calculating the radiation dose due to the exposure of radon, thoron and their progeny, the equilibrium factors should be determined separately for individual dwelling.

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