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Thoron Equilibrium Factor Observed around Chhatrapur Placer Deposit, a High Background Radiation Area in Odisha, India

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In thoron (^{220}Rn)-prone areas, the contribution from thoron (including progenies) can be equal to or exceed that from radon (^{222}Rn) during radiation exposure. The dose estimation is practically performed using the thoron concentration and thoron equilibrium factor. In the present study, thoron equilibrium factors were determined from direct measurements of thoron and its progeny concentrations for dwellings in the high background radiation area of Odisha, India. The results show that the equilibrium factor has a seasonal variation, with a minimum in winter and a maximum in summer. The frequency distributions exhibit log normality with geometric means of 0.025, 0.044, and 0.051 in winter, rainy, and summer-seasons, respectively. The annual average of 0.04 is observed to be in the order of the United Nations Scientific Committee on the Effects of Atomic Radiation recommended value (i.e., 0.02).

Key words: radon, thoron, thoron progeny, equilibrium factor, inhalation dose, high background radiation area

1. Introduction

Radon isotopes (radon (^{222}Rn) and thoron (^{220}Rn)), chemically inert radioactive gases that are generally abundant in the living environment, have an important role in radiation exposure among natural radiation sources. According to the estimation made by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[1)], the dose received from inhalation of radon, thoron, and their progenies dominates approximately 50% of the global average annual radiation dose (1.2 mSv vs. 2.4 mSv). In particular, the contribution of the dose from radon is estimated to be up to 90% and the dose from thoron is small as thoron concentration is lower due to the difference in their half-lives (3.8 d for radon and 55.6 s for thoron). However, recent surveys performed in the past decade have revealed the existence of a thoron-prone area where doses from thoron can be equal to or exceed those from radon[2-7)]. In addition, some

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researchers[8-9] proposed a dose conversion factor for thoron, which was based on the latest human respiration and bio-kinetic models and was three times higher than the dose conversion factor adopted by the UNSCEAR. Thus, exposure to thoron has been recognized as an important factor.

Thoron progenies are responsible for lung exposure to thoron in the dose estimation. However, the measurement of thoron progeny concentration is generally complex compared to that of thoron gas concentration using the passive technique. In sequence, the measured thoron concentration is multiplied by the thoron equilibrium factor to estimate the equilibrium equivalent thoron concentration as the thoron progeny concentration[1, 10-15]. It represents the degree of equilibrium between thoron and its progeny (^{212}Pb and ^{212}Bi)[16]. The UNSCEAR report determined a typical value of 0.02 for the thoron equilibrium factor[1]. In contrast, thoron equilibrium factors compiled from regional surveys with long-term measurements differed by three orders of magnitude among the dwellings, although representative values (arithmetic or geometric mean) were in the order of the UNSCEAR computed value[17, 18]. These long-term measurements were generally performed over several months and were not completed within a year[19-22].

We conducted a thoron and its progeny survey of approximately 100 dwellings in the southeastern coastal area of India from 2010 to 2011[5, 23-24]. The meassurements were obtained for approximately four months and repeated three times. In the present study, based on the database of thoron and its progeny concentrations, thoron equilibrium factors were calculated and their variation over seasons was examined.

2. Materials and methods

2.1. Outline of location and conducted survey

The thoron and its progeny survey was conducted in the Chhtrapur placer deposit area along the southeastern coast of Odisha (Orissa), India (location map present in Omori *et al.*[5]). The absorbed dose rate in air in the area is up to approximately 5000 nGy h^{-1}; therefore the area is identified as a high background radiation area[25, 26]. For the survey, 106 dwellings (100 cement constructions and 6 mud constructions) were selected from 12 villages in the study area. A pair of thoron and its progeny detectors were installed on the wall (a few centimeters from the wall surface) or the middle of the room in each dwelling. The exposure period was set to be approximately four months, and the measurements were repeated three times (first period: mid-October 2010–early February 2011 (winter); second period: mid-February–early June 2011 (summer); and third period: mid-June–mid-October 2011 (rainy season)) to examine the differences throughout the year. Details of the survey can be found in Omori *et al.*[5]

2.2. Measurement of thoron concentration

The thoron concentrations were measured using solid-state nuclear track detectors enclosed in diffusion chambers, RADUET (Radosys Co. Ltd., Hungary). A combination of chambers with different air exchange rates can measure radon and thoron concentrations separately[27]. A solid-state nuclear track detector CR-39 with dimensions of 10 mm × 10 mm (Radosys Co. Ltd., Hungary) was used to record tracks of alpha particles from radon, thoron, and their progeny. The RADUETs were calibrated using radon and thoron chambers established by the National Institute of Radiological Sciences (NIRS; currently National Institutes for Quantum Science and Technology)[28, 29].

The RADUET was positioned on the wall or in the middle of the room in each dwelling to measure the average thoron concentration over approximately four months indoors. After the exposure was completed, the RADUETs were processed in NIRS for chemical etching of the CR-39 chips, counting the number of tracks registered on the chips, and evaluating thoron concentrations. The CR-39 chips were etched in 6.25 M NaOH solutions at 90 °C for 6 h. The number of tracks formed on the chips was counted using an automatic reading system, Radometer2000 version 1.43 (Radosys Co. Ltd., Hungary), and it was divided by the analyzed area to determine the track density. Thoron concentration was estimated from the exposure period, track density, and calibration factors for thoron and radon exposure[5].

2.3. Measurement of thoron progeny concentration

Thoron progeny concentrations were measured using a deposition-based technique[30, 31]. The thoron progeny detector used in the present study has the function of alpha-energy discrimination to detect alpha particles from one of the thoron progenies ^{212}Po (8.8 MeV) only. CR-39 chips with dimensions of 10 mm × 10 mm (Nagase Landauer, Ltd., Japan) were mounted on a stainless steel plate and covered with two-layered Mylar films. The thickness of the Mylar films was adjusted such that alpha particles from ^{212}Po could pass through the film and reach CR-39.

The thoron progeny detector was positioned on the wall or in the middle of the room together with RADUET over approximately four months. After the exposure was completed, the thoron progeny detectors were processed in NIRS for chemical etching of the CR-39 chips, counting the number of tracks registered on the chips, and evaluating the thoron progeny concentrations. The CR-39 chips were etched in 6 M NaOH solutions at 60 °C for 24 h. The number of tracks formed on the chips was counted using image capture and analysis. In the

Table 1. Statistical values of thoron equilibrium factor during the measurement periods

Season	N[a]	Thoron equilibrium factor		
		AM ± SD[b]	GM (GSD)[b]	Range
Winter	62	0.030 ± 0.019	0.025 (1.89)	0.006 - 0.087
Summer	76	0.066 ± 0.059	0.051 (1.99)	0.016 - 0.42
Rainy	41	0.056 ± 0.044	0.044 (2.05)	0.012 - 0.24

[a] N means the number of dwellings for analysis.
[b] AM, SD, GM and GSD mean arithmetic mean, standard deviation, geometric mean and geometric standard deviation (no unit), respectively.

image capture, ten photographs (approximately 17.7 mm^2 in total) of the formed tracks were taken manually for every etched chip at 100× magnification with an optical microscope and camera (Model LBV-A570, KENIS Ltd., Japan). Image analysis was performed to count the tracks captured on the digital photographs using CellProfiler 2.0, software developed by Carpenter *et al.*[32] The equilibrium equivalent thoron concentration was estimated from the exposure period, track density, and a calibration factor for thoron progeny exposure[5].

2.4. Analysis

Thoron equilibrium factor (F_{Tn}) was calculated from the thoron concentration (C_{Tn}) and equilibrium equivalent thoron concentration ($EETC$) determined by the RADUET and thoron progeny detector, respectively, as follows:

$$F_{\mathrm{Tn}} = \frac{EETC}{C_{\mathrm{Tn}}}. \quad (1)$$

At three of the 106 dwellings investigated, the RADUET and thoron progeny detector were deployed in the middle of the room. Data obtained in these cases were excluded from the analysis.

Statistical and regression analyses were performed using the software OriginPro 2020 (OriginLab Corporation). Statistical significance was set at a level of 0.05.

3. Results and discussion

Thoron equilibrium factors were evaluated successfully in 62, 76, and 41 dwellings in the first (winter), second (summer), and third (rainy season) periods, respectively. In 24 dwellings, the total measurement period covered a year. Thoron equilibrium factors for some dwellings were not evaluated because the thoron concentrations were lower than the detection limits for the first and second periods. The smaller number of samples in the third period was because of the smaller number of investigated dwellings.

Figure 1 shows the distributions of the thoron equilibrium factor in the form of cumulative probability

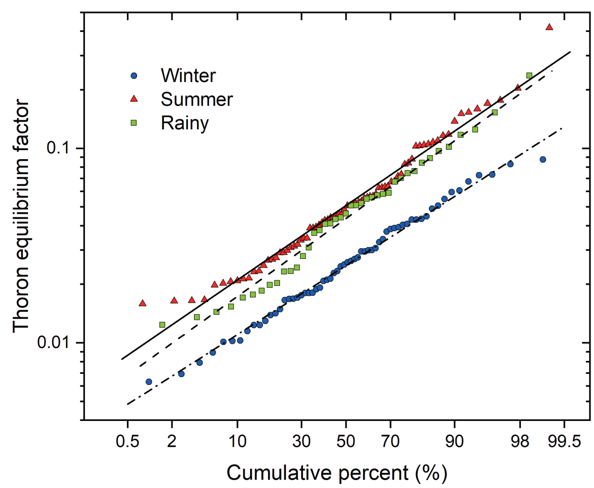

Fig. 1. Distribution of thoron equilibrium factor in the form of cumulative probability on a logarithmic scale. Fitted (dotted, dashed, and solid) lines are drawn by estimating ideal log-normal distributions from the geometric mean and geometric standard deviation of the thoron equilibrium factor.

on a logarithmic scale. The Shapiro–Wilk test for the equilibrium factor after logarithmic transformation suggests that it had a lognormal distribution in all periods (p-values were 0.79, 0.14, and 0.59 for datasets in winter, summer, and rainy seasons, respectively). This was evident from the regression analysis for the plots in Figure 1, which shows that a line drawn under an ideal lognormal distribution fitted well.

The statistic results for the thoron equilibrium factors are summarized in Table 1. In the summer period, the thoron equilibrium factors were distributed in a range of 0.016–0.42 with arithmetic and geometric means of 0.066 and 0.051, respectively. The thoron equilibrium factors differed by one order of magnitude in the minimum and maximum values. A similar characteristic was found in the dataset for the rainy period, where the equilibrium factors were distributed in the range of 0.012–0.24 with arithmetic and geometric means of 0.056 and 0.044, respectively. In contrast, all the equilibrium factors in the winter period were in the range of 0.006–0.087, with arithmetic and geometric means of 0.030 and 0.025,

Table 2. Summary of seasonal variations in thoron equilibrium factors

Location	Thoron equilibrium factor Arithmetic mean ± standard deviation (range)				Detector position (distance from wall)
	Dry season		Rainy season		
Lolodorf, Cameroon[6]	0.25 ± 0.05 (0.02 - 0.85)		0.09 ± 0.01 (0.01 - 0.21)		20 cm
	Winter	Summer	Rainy	Autumn	
Uttarkhand, north India[33]	0.04 (0.01 - 0.80)	0.03 (0.01 - 0.27)	0.04 (0.01 - 0.63)	0.04 (0.01 - 0.53)	10 cm
	Winter	Summer	Rainy		
Uttarkhand, north India[34]	0.06 ± 0.05 (0.01 - 0.21)	0.06 ± 0.05 (0.01 - 0.31)	0.09 ± 0.06 (0.02 - 0.36)		10 cm
Uttar Pradesh, north India[35]	0.01 ± 0.01 (0.005 - 0.04)	0.01 ± 0.01 (0.005 - 0.036)	0.01 ± 0.01 (0.005 - 0.032)		30 cm
Punjab, north India[36]	0.04 (0.01 - 0.11)	0.03 (0.01 - 0.08)	0.04 (0.01 - 0.11)		30 - 50 cm
Punjab, north India[37]	0.05 ± 0.03	0.05 ± 0.04	0.04 ± 0.03		20 cm
Jammu and Kashmir, north India[38]	0.03 ± 0.02 (0.01 - 0.09)	0.04 ± 0.08 (0.005 - 0.09)	0.03 ± 0.02 (0.01 - 0.07)		10 - 20 cm
Odisha, east India (Present study)	0.03 ± 0.02 (0.006 - 0.09)	0.07 ± 0.06 (0.02 - 0.42)	0.06 ± 0.04 (0.01 - 0.24)		On wall

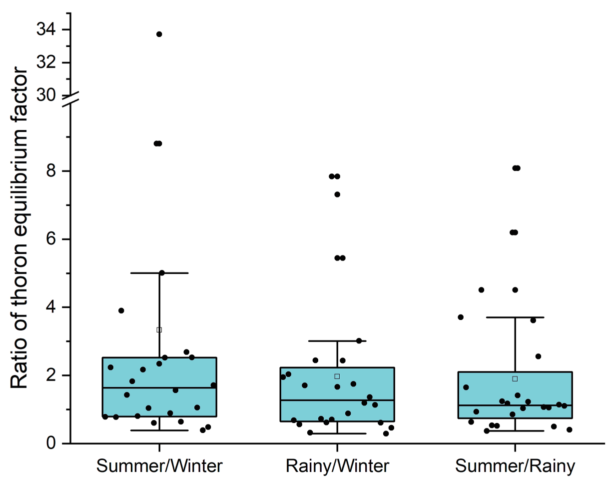

Fig. 2. Distributions of thoron equilibrium factor ratios among summer, rainy, and winter seasons. Boxes are bounded at 25 percentile value, median value, and 75 percentile value of the ratios from lower to upper, respectively. Open squares represent the arithmetic mean.

respectively. Compared with the summer and rainy periods, the maximum thoron equilibrium factor was smaller by one order of magnitude in the winter period. The annual means for the 24 dwellings were in the range of 0.01–0.16 with an arithmetic mean of 0.04.

The thoron equilibrium factors reported in the present study agreed with those summarized from the results of regional surveys by Hosoda *et al.*[17] and Chen *et al.*[18] They reported wide ranges of thoron equilibrium factors from 0.0001 to 0.1. Similarly, the present study reports a wide range of thoron equilibrium factors. However, they were less than or in the order of the value recommended by the UNSCEAR (0.02)[1] for 100%, 80%, and 90% of the cases in which thoron equilibrium factors could be determined in winter, summer, and rainy seasons, respectively (Fig. 1).

The thoron equilibrium factors showed seasonal variation. In other words, those in winter appeared lower than those in the other seasons. To confirm the seasonal variation, ratios of the equilibrium factors were calculated between the seasons for the 24 samples, that is, ratios from summer to winter, rainy to winter, and summer to rainy. Figure 2 shows the distributions of the ratios of the thoron equilibrium factors between the seasons. The thoron equilibrium factors in the summer and rainy seasons relative to those in the winter season were higher than unity for most samples. In other words, the thoron equilibrium factors in winter were generally lower than those in the other seasons, which is expected from the results in Figure 1. Similarly, the thoron equilibrium factors in the rainy season were slightly lower than those in the summer.

Table 2 summarizes the seasonal variations in the thoron equilibrium factor reported in previous studies[33-38] and the present study. This clearly shows that the thoron equilibrium factors do not exhibit typical seasonal variations. In detail, the equilibrium factors were higher in the colder seasons in some cases, higher in the warmer seasons in other cases, and constant throughout the seasons in other cases. Ideally, the thoron equilibrium factor decreases with an increase in the ventilation rate[39, 40]. The equilibrium factor is expected to be higher in colder seasons due to the poor ventilation of the room air. However, such seasonal variation was rarely observed in previous studies[33-38] and in the present study. This is partly because the behavior of thoron in room air is very complicated due to its short half-life. In particular, in the colder season, room air does not mix well due to poor ventilation. In this situation, the spatial distribution of thoron concentration becomes more heterogeneous, and the concentration gradient increases significantly at locations close to the thoron source (i.e., walls, floors, and ceilings). This phenomenon reduces the thoron equilibrium factor if the thoron progeny concentration is constant. To better understand seasonal variation in the thoron equilibrium factor, together with thoron and its progeny concentrations, aerosol concentration should be measured, which has a role in changes in the thoron equilibrium factor.

4. Conclusion

The main findings of the present study are summarized as follows:

- Thoron equilibrium factors followed a log-normal distribution with geometric means of 0.025, 0.044, and 0.051 in the winter, rainy, and summer seasons, respectively;
- In 80–100% of the cases for each of the three seasons, thoron equilibrium factors showed values lower than or in the order of the value recommended by UNSCEAR (0.02)[1];
- Thoron equilibrium factors showed seasonal variation with a minimum in winter and maximum in summer;
- Typical seasonal variation for the thoron equilibrium factor could not be determined based on comparisons with previous studies and the present study.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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