Study of indoor ²²²Rn and ²²⁰Rn progeny using augmented passive detectors

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ABSTRACT

Background: Prognostic models and specific data are the basics of radiation dosimetry. Inhalation is the major pathway of radiation exposure to man. Precise data of progeny concentration status is important for all dose assessments owing to Radon (²²²Rn) and Thoron (²²⁰Rn). **Materials and Methods:** The measurements of unbound (unattached) and bound (attached) short lived Radon and Thoron daughters were carried out in selected dwellings of coastal regions of Thiruvananthapuram district, Kerala. For the precise assessment of fraction of bound and unbound fraction of radon / thoron progeny, progeny sensors with wire mesh capped progeny sensors were used. The method makes use of radon thoron progeny discrimination using proper barriers and wire mesh helps to segregate bound against unbound fractions of progeny. **Results:** Overall (bound and unbound) equilibrium equivalent concentrations of ²²²Rn (EECRA+U) and similar quantity for ²²⁰Rn concentration (EECTA+U) were found to differ from 6.31± 0.9 to 7.98 ± 1.1 Bqm⁻³ and 0.35 ± 1.1 to 0.61 ± 0.1 Bqm⁻³, respectively. **Conclusions:** The volume specific quantities of bound progeny nuclides for both Radon and Thoron were found superior than the unbound daughter nuclides in the residences of the study area. The unattached (unbound) equilibrium equivalent concentrations of ²²²Rn and ²²⁰Rn did not demonstrate any notable relationship with ventilation conditions. It was found that there is a positive relationship between the progeny attachment rate and aerosol concentration for radon.

INTRODUCTION

Following the decay of ²²²Rn gas, the subsequently produced alpha active progeny (²¹⁸Po, ²¹⁴Po, and ²¹⁰Po) may attach to aerosols and dust granules in the atmosphere. A considerable fraction remains unattached. The free or unbound radon progeny have an important role towards the dose to human respiratory organs through inhalation of radon daughters. The southern coastal region of India is known to have rich deposits of thorium with slightly higher concentrations of uranium [1]. Therefore, there exists a unique situation where radon and thoron gases together contribute to the inhalation dose. Hence, in this region along with radon, alpha active thoron progeny (²¹⁶Po, ²¹²Bi, and ²¹²Po) are also present [2, 3]. In spite of their dosimetric importance, the data on unbound fraction of progenies of radon and thoron are scarce owing to their relatively low levels and difficulty in assessment.

Radon (²²²Rn) and Thoron (²²⁰Rn) existing inside dwellings atmosphere is a likely foundation of inhalation dose and is typically contributed by breathing in their progenies [4, 5]. The progenies are electrically charged at the time of decay and they get sticked to surfaces or aerosols existing in air,

classified as bound progeny. Generally, radionuclides of 0-3 nm in size are unbound while the bound radionuclides have 100 – 500 nm size [6-8]. It is an established fact that the inhalation dose imparted to the target tissues in the bronchial epithelium of the human lungs are predominantly contributed by the unbound fractions [8, 9]. The radon inhaled and its alpha releasing airborne daughter products may be one the root cause for lung carcinoma owing to the radiation dose absorbed in trachea bronchial epithelium [10-12]. As per a study in Korea, lung malignancy results in maximum mortality of 34.4 per 100,000 individuals [13, 14]. According to the WHO Report and many other research findings, inhalation of radon and progeny of it is accounted as the second important reason for lung cancer after smoking [15-19].

For a precise measurement of inhalation radiation dose, the data of the concentrations of unbound and bound progenies in living space is important. On the whole, these natural or human caused airborne contaminants are originally in a microscopic state. Removal of these radionuclides takes place by radioactive decay or by elimination processes like ventilation and plate out [20, 21]. During inhalation, the radionuclides in the inhaled air may be placed in different respiratory tract regions [22]. The deposition

of the progeny is controlled by the physical characteristics of the aerosols in indoor atmospheres [23, 24].

The present study makes use of Deposition based Radon Progeny Sensor (DRPS) and Deposition Thoron Progeny Sensors (DTPS) for the discrimination of progenies of the gases. While unbound and bound ^{222}Rn and ^{220}Rn progeny assessment was made using the wire mesh capping. These are indigenously developed sensors at Bhabha Atomic Research Center (BARC), India for direct measurement of progeny concentrations [25]. The study was conducted in the selected dwellings along the coastal region of the capital district Thiruvananthapuram of the state of Kerala in India. While DRPS and DTPS measure the progeny concentrations of radon and thoron, wire mesh capped direct radon and thoron progeny sensors were used to estimate the unbound progeny levels of the progenies. The theory of the DRPS / DTPS are detailed at large elsewhere [24]. The area selected for the study is not a high background radiation area. However, some parts of the region of investigation are known to have radioactive pockets [25]. The objective was to make the assessment of unbound and bound progeny levels experimentally using DRPS/DTPS fixed with wire mesh in indoor environment in the High Background Radiation Area (HBRA) in the coastal regions. The study is unique as the region has elevated levels of radon as well as thoron.

Solid state track detection is a simple, direct and reasonably accurate method for radon, thoron and progeny determination. There are several passive detector systems with capability to discriminate radon and thoron gases. Based on these detector systems numerous reports on various aspects of the gas and progeny assessment are available in literature over the last decades [26-28]. DRPS and DTPS are modified solid sensors specifically designed for the estimation of radon and thoron gas progenies. There are modified designs of these sensors for the determination of unbound and bound fractions of the alfa active progeny of radon and thoron gases [25, 29, 30].

MATERIALS AND METHODS

Solid state Nuclear Track Detectors are being used widely for radon and thoron studies [31-34]. DTPS was fixed with passive particle track detector (LR-115 Type II pelliculable films made by Kodak Pathe, France) mounted with aluminized mylar absorbers of 50 μm thickness on them. The LR 115 detector films selectively record tracks of 8.78 MeV alpha radiations from the radioactive decay of ^{212}Po atoms deposited on the absorber surface. Alpha particles originating from the progeny of radon and thoron deposited on the absorber surface will not be able to record tracks

in the detector as their energy is degraded below 4 MeV (figure 1a) so that their alpha tracks are not recorded in the LR-115 films. DRPS also has the detector film strips of LR-115 type II fixed with aluminized mylar and cellulose nitrate absorbers with thickness 37μm to record tracks mainly 7.69 MeV alpha particle originating from ^{214}Po (radon progeny) [25, 29]. In both DTPS and DRPS, the absorbers help to discriminate the alpha radiations of different energies originating from the plated-out progeny of radon and thoron (figure 1b).
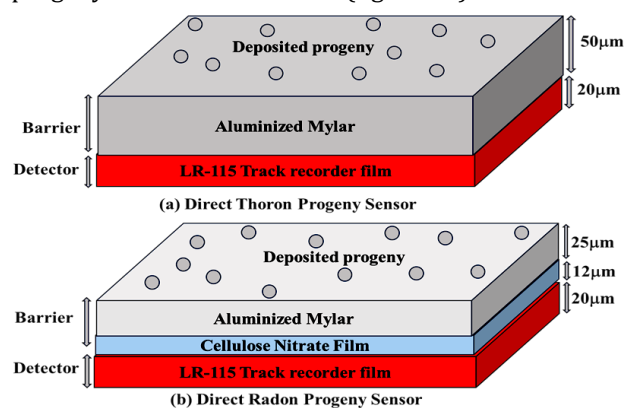

Figure 1. Direct thoron progeny sensor **(a)** and direct radon progeny sensors **(b).**

Higher risks of lung cancer are related with inhalation and deposition of short-lived ultrafine (less than 2nm) unbound radon progeny in the lung [30]. In order to segregate unbound and bound fractions of progenies of these radioactive gases in the atmosphere, DRPS/DTPS fixed with wire-mesh were used. It consists of DRPS/DTPS masked with a wire screen of mesh size 79 cm^{-1} having wire diameter 0.005 cm. The wire mesh fixed at an optimized distance on the detector helps to trap the fine fraction and the tracks registered in the detector is negligible [31]. The portion of the unbound progenies stuck on to the wire–mesh, and the bound fraction of the progeny atoms gets plated out on the DRPS/DTPS to get registered [32]. Hence, tracks of alpha radiations on the capped sensors give a quantity of the bound portion of progeny level. A schematic drawing of wire-mesh fixed DTPS/DRPS along with the absorbers is shown in figure 2.
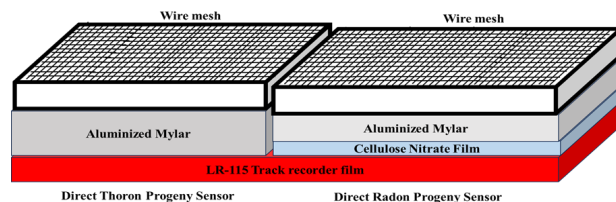

Figure 2. Wire-mesh capped DTPS and DRPS.

Exposure of detector films fixed with absorbers and wire mesh record tracks of alpha radiations from the progenies. A measure of equilibrium equivalent thoron concentration (EETC) of the thoron daughter products can be determined right from the track density of alpha radiations on the detector films in

the DTPS. The absorber in the DTPS is of relatively thicker (50μm) and prevents radon progeny to pass through it. For the assessment of equilibrium equivalent radon concentration (EERC), the alpha radiations from both radon and thoron progenies pass through the absorber (37μm) since the energy of ^{212}Po (thoron daughter) is more than that of ^{214}Po (radon progeny). Hence to determine EERC, the alpha tracks created by thoron daughter products are disregarded as calculated from DTPS, using the equation 1 [31, 35].

Tracks of radon progeny = Total tracks on DRPS - $\frac{\eta_{RT}}{\eta_{TT}}$×Tracks on DTPS   (1)

Where η_{RT} (0.01±0.0004) and η_{TT} (0.083±0.004) are the track registration efficiencies of thoron progeny in DRPS and DTPS, respectively. To calculate EETC and EERC (in Bq m^{-3}) the equations 2 and 3 were used [25, 29].

$$EETC = \frac{Total\ Track\ density\ on\ DTPS}{K_T \times Exposure\ Period} \quad (2)$$

$$EERC = \frac{Total\ Track\ density\ due\ to\ radon\ on\ DRPS}{K_R \times Exposure\ Period} \quad (3)$$

In these equations, K_T and K_R are calibration factors for DTPS and DRPS, respectively. These factors for DTPS and DRPS are 0.94±0.057 tracks cm^{-2} d^{-1}/EETC (Bq m^{-3}) for DTPS and 0.09±0.0036 tracks cm^{-2} d^{-1}/EERC (Bq m^{-3}) for DRPS [30, 31].

After the exposure of progeny detectors and wire mesh capped detectors for the specified period of 3 months, in twenty-seven sites in five locations, all the LR-115 track detectors were retrieved. The long-term exposure helps us to obtain accurate time averaged assessments of gas progeny. After the preferential chemical etching to reveal the alpha tracks in the detectors and scanning, the average track densities were obtained on each LR-115 detector film. Using the track densities, equilibrium equivalent concentrations of radon and thoron were obtained using the respective calibration factors. In the similar manner, track densities on the detector films fixed in the DRPS and DTPS capped with wire-mesh were used to determine the equilibrium equivalent concentrations of unbound and bound portions of radon and thoron. Using these data radon and thoron daughter attachment rate to the aerosol particles was calculated. Concentration of aerosols using unbound fraction of Radon progeny was also determined. Using these data, equilibrium equivalent progeny concentrations (EERC$_A$ and EERC$_{UN}$ for radon and EETC$_A$ and EETC$_{UN}$ thoron) were determined for bound and unbound fractions of the progeny.

Determination of rate of progeny attachment (X)

The rate of attachment of the ^{222}Rn and ^{220}Rn daughter products to the aerosol particulates in the indoor environment was calculated using the equations 4 and 5 [31, 35].

$$R_X = \frac{EERC_A}{EERC_{UN}}(l_1 + 1) \quad (4)$$

$$T_X = \frac{EETC_A}{EETC_{UN}}(l_2 + 1) \quad (5)$$

Where; EERC$_A$, EETC$_A$ are the concentrations of attached ^{222}Rn and ^{220}Rn progenies. EERC$_{UN}$, EETC$_{UN}$, are the concentrations of unattached ^{222}Rn and ^{220}Rn progenies. l_1 and l_2 are the decay constants of ^{222}Rn and ^{220}Rn progeny respectively.

Aerosol Concentration (Z)

The concentration of aerosol in the atmosphere and the rate of attachment of the progeny are directly related. Using the unbound fraction (f) of Radon progeny, aerosol concentration (Z) was determined based on the equation 6 [6].

$$Z = \frac{414}{f} \quad (6)$$

Statistical analysis

This manuscript reports the measurement of effective equivalent concentrations of radon thoron, bound and unbound fractions of progenies, attachment rate and aerosol concentrations in air have been determined. All these parameters have been estimated based on the track densities recorded on the alpha detector films. Track counting was performed in a minimum of 100 fields with a spark counter specifically designed for the purpose (manufactured by Polltech Instruments Pvt. Ltd., India) [36, 37]. The average and standard deviation for each film were calculated using excel program. The level of significance was set to be 0.05.

RESULTS

The track densities on the detector films were used to estimate EERC and EETC, bound and unbound fractions of progenies, attachment rate and aerosol concentrations in air. Tables 1 and 2 describe all the parameters determined for radon, thoron and their progeny for the five locations of study. The uncertainties shown are the statistical counting errors estimated during scanning. The results indicate that there is no appreciable variation in the total (attached + unattached) EERC between the regions. While we examine the bound and unbound fractions of progeny levels there are appreciable dissimilarities. Since the data were non-normally distributed, we did Mann-Whitney U test and found to have p-value < 0.05. Attachment rate of progenies to aerosols and aerosol concentrations in all the five locations were also determined.

Figure 3(a) is the graphical representation of variation of aerosol concentration, z with radon progeny attachment rates, R$_X$ and 3(b) is the similar representation for thoron progeny attachment rates,

T_X at all the sites in five locations. It can be seen that the two parameters, the aerosol concentration and attachment rate of radon on aerosols bear a linear correlation (R = 0.913, P<0.05).

Meanwhile, no such relation was observed for thoron progeny. In the case of thoron progeny, attachment rates had a wider range as compared with the radon progeny attachment rates. Unlike radon progeny, thoron progeny gets attached even at lower concentrations of aerosols.

Figure 4(a) shows the variation of radon attachment fractions (f_A^R) and progeny attachment rates for radon (R_X) and 4(b) is the variation of thoron attachment fractions (f_A^T) and thoron progeny attachment rate (T_X) for the five locations. The mean rate of attachment of ^{222}Rn, (R_X) and that of ^{220}Rn, (T_X) progeny was found to vary from 1.27± 0.3 to 3.49 ± 0.7 s^{-1} and from 1.34± 0.2 to 13.8 ± 2.6 s^{-1}, respectively. The results of the experiments showed an exponential relation between the attachment rate (R_x and T_x) and bound portion of ^{222}Rn (f_A^R) and ^{220}Rn (f_A^T) progeny (figure 4).

The over-all (attached plus unattached) equilibrium equivalent concentration of ^{222}Rn (EECR$_{A+U}$) were between (6.31±0.9) and (7.98±1.1) Bq/m^3. Over-all (attached plus unattached) equilibrium equivalent concentration of ^{220}Rn (EECT$_{A+U}$) were varied from (0.35 ± 0.1 to 0.61± 0.1) Bq/m^3, respectively. The estimated average indoor aerosol concentration (Z) in the five locations were found to vary from 938 ± 266 to 1856 ± 436 cm^{-3}.

Table 1. Average indoor Radon progeny parameters for the five locations.

Location	Varkala	Kovalam	Shangumugam	Vizhinjam	Poovar
Lat-Long coordinates	8°44.5'N 76°42.1'E	8°23.0'N 76°58.9'E	8°28.5'N 76°54.9'E	8°22.7'N 76°59.6'E	8°20.9'N 77°01.7'E
No. of sites	5	5	6	5	6
EERC$_{(A+U)}$ (Bq/m^3)	7.98 ± 1.1	7.65 ± 1.1	6.55 ± 0.9	6.31± 0.9	7.61± 1.1
EERC $_A$ (Bq/m^3)	6.21± 0.9	4.41 ± 0.7	3.66 ± 0.7	4.53 ± 0.8	5.19 ± 0.8
EERC$_{UN}$ (Bq/m^3)	1.78 ± 0.2	3.25 ± 0.3	2.89± 0.2	1.78± 0.2	2.42± 0.2
Attached fraction (f_A^R)	0.77± 0.1	0.57± 0.1	0.55± 0.1	0.71± 0.1	0.68± 0.1
Attachment rate (s^{-1})	3.49 ± 0.7	1.35± 0.4	1.27± 0.3	2.55 ± 0.7	2.15 ± 0.6
Aerosol Conc. (cm^{-3})	1856 ± 436	975 ± 264	938 ± 266	1468 ± 316	1302± 296

(a) Attachment rate of radon progeny
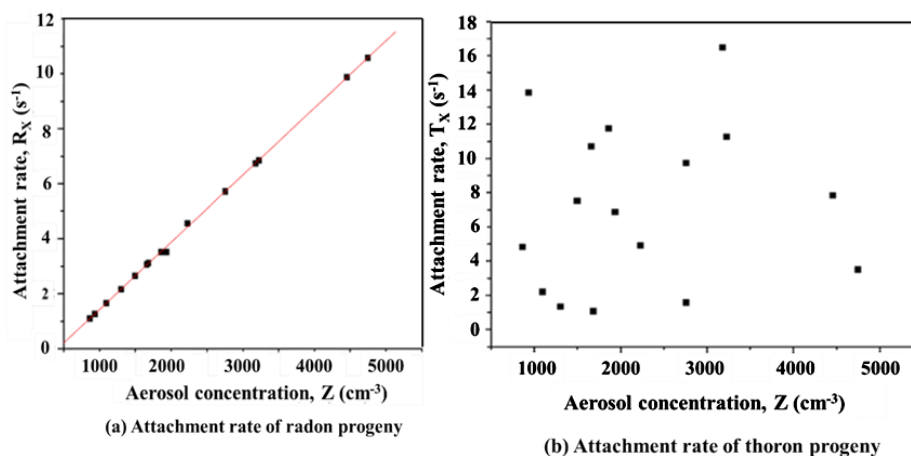
(b) Attachment rate of thoron progeny

Figure 3. (a) Variation of aerosol concentration (Z) and radon progeny attachment rates (RX) and **(b)** variation of aerosol concentration (Z) and thoron progeny (TX).

Table 2. Average indoor thoron progeny parameters for the five locations.

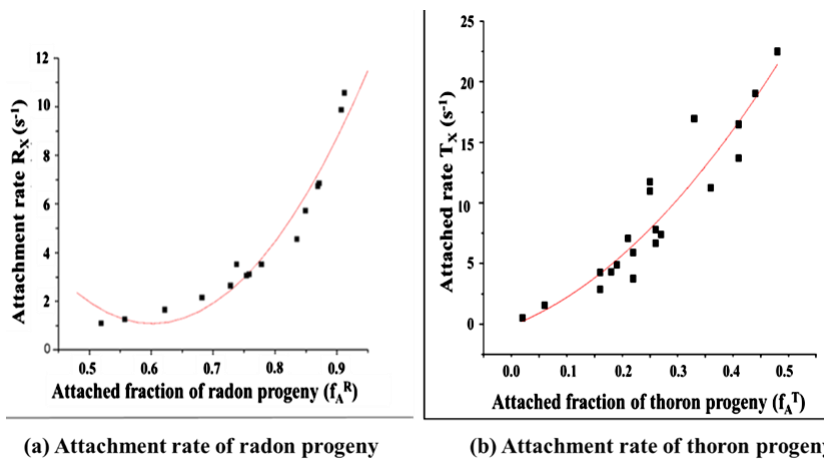| Location	Varkala	Kovalam	Shangumugam	Vizhinjam	Poovar
Lat-Long coordinates	8°44.5'N 76°42.1'E	8°23.0'N 76°58.9'E	8°28.5'N 76°54.9'E	8°22.7'N 76°59.6'E	8°20.9'N 77°01.7'E
No. of sites	5	5	6	5	6
EETC$_{(A+U)}$ (Bq/m^3)	0.38 ± 0.1	0.35 ± 0.1	0.61 ± 0.1	0.41 ± 0.1	0.38 ± 0.1
EETC $_A$ (Bq/m^3)	0.27± 0.06	0.16± 0.04	0.4± 0.09	0.11± 0.03	0.07± 0.06
EETC$_{UN}$ (Bq/m^3)	0.11± 0.02	0.19± 0.02	0.21± 0.02	0.29± 0.02	0.31± 0.03
Equi. Factor of Th (F_{Th})	0.027± 0.08	0.034± 0.09	0.029± 0.08	0.032± 0.08	0.021± 0.06
Attached fraction (f_A^T)	0.71± 0.1	0.46± 0.08	0.65± 0.09	0.28± 0.04	0.19± 0.02
Attachment rate (s^{-1})	13.8 ± 2.6	4.82 ± 0.6	10.71± 2.2	2.21± 0.3	1.34± 0.2

Figure 4. Comparison of attachment fractions (fAR and fAT) and progeny attachment rates for radon (RX) and thoron progeny (TX).

(a) Attachment rate of radon progeny   (b) Attachment rate of thoron progeny

DISCUSSION

The graphical representations shown as figure 3 indicate a strong linear relationship between the aerosol concentration and attachment rate of radon on aerosols (R=0.913, P<0.05). The variation of aerosol concentrations could be attributed to the different geographical nature of the locations with proximity to the Arabian sea. When the concentration of aerosols in the indoor air rises, the attachment rate of the progenies to the aerosols enhances [38, 39]. It is a well-accepted fact that burning coal and wood increase the aerosol concentration (Z, cm^{-3}) in the indoor atmosphere to enhance the process of progeny attachment [40]. Hence, a positive relation is probable amongst aerosol concentration (Z, cm^{-3}) and attachment rate (R_X, s^{-1}). Nevertheless, the such a relation was not seen between attachment rate (T_X, s^{-1}) and aerosol concentration (Z, cm^{-3}). The greater part of the unbound radon daughter products is attached in the respiratory tract during breathing, whereas 80% of the bound radon progeny are exhaled without getting attached in the lung tissues [32]. The unbound activity sums up to about 10% of the overall radioactivity. But it is considered to yield about 50% of the total radiation dose to the lung tissues [40]. Measurements of the bound and unbound fractions are indispensable for the precise evaluation of inhalation dose as the models require parameters of activity distributions of thoron and radon progeny. Enhancement in the size and concentration of the aerosols inclines to upsurge the rate of attachment due to more probable interactions between the aerosols and progeny and thus reducing the unbound fraction. The findings of the present research are in good agreement with the results of similar studies held in India [41-43].

CONCLUSION

Bound fractions of progenies for both ^{222}Rn and ^{220}Rn were much larger than the unbound fractions of progenies in the studied area. Equilibrium equivalent concentrations of ^{222}Rn and ^{220}Rn progeny were higher in poorly ventilated condition as compared to the partially ventilated or well-ventilated conditions. The unbound equilibrium equivalent concentrations of ^{222}Rn and ^{220}Rn did not demonstrate any notable relationship with ventilation conditions. There was a positive relationship between the progeny attachment rate and aerosol concentration for radon. The results of the measurements show a wide variation of progeny levels and their attachment rates as these parameters depend on environmental features and conditions. Therefore, site-specific dose estimations are essential for an accurate risk and health impact modeling.

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