Attached Progeny Profile in a High Level Radon-Thoron Coastal Environment

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Abstract

Wide inconsistency in indoor radon and thoron levels has been reported in the Monazite bearing densely populated regions in the coastal belt of Kerala, India. Inhalation doses are predominantly due to these gas progenies and the unattached fraction is significant in determining radiation dose to the human lungs. The attached and unattached radon and thoron concentrations have been determined using direct radon (222Rn) and thoron (220Rn) progeny sensors (DRPS/DTPS) in the Neendakara and Chavara hamlets of Kollam district, Kerala. The equilibrium equivalent concentration of the unattached fraction of 222Rn and 220Rn (EERC\textsubscript{U} and EETC\textsubscript{U}) was found to vary from 0.5 to 2.4 Bqm\textsuperscript{-3} and 0.1 to 0.2 Bqm\textsuperscript{-3}, respectively. The concentrations of the attached fraction of 222Rn were found to be greater than the unattached fraction. The attached fraction of 220Rn progeny was in the range 1.01 to 3.8 Bqm\textsuperscript{-3}. The total (attached + unattached) equilibrium equivalent 222Rn concentration (EERC\textsubscript{A+U}) and total (attached + unattached) equilibrium equivalent 220Rn concentration (EETC\textsubscript{A+U}) were found to vary from 8.8 to 22.5 Bqm\textsuperscript{-3} and 1.1 to 4 Bqm\textsuperscript{-3}, respectively. Annual effective dose rates were also determined.

Keywords: Unattached fraction, progeny, DTPS, DRPS

Introduction

Monazite sand along the south coast of Kerala, India typically contains thorium oxide (~9%) and uranium oxide (0.35%) along with rare earth minerals. Mixed radon-thoron environment existing in this thickly populated high background radiation area is unique. In an atmosphere of 222Rn, 220Rn and their short lived decay products, the major part of radiation dose to human lung comes from the inhaled and subsequently deposited radon and thoron progeny\textsuperscript{1-3}. The progeny are present in the environment in both attached and unattached form. The decay products of 222Rn and 220Rn are electrically charged and tend to attach themselves to aerosols and dust particles in the air. The particle size range of attached and unattached radionuclides is 100-500nm and 3nm respectively\textsuperscript{4-5}. The unattached fraction is significant in determining the dose to the human lung\textsuperscript{6-9}.

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The attached fraction passes the upper respiratory tract and is more likely to leave alveoli during exhalation. But major part of the unattached fraction passes the upper respiratory tract and gets deposited in the alveoli. The unattached fraction is absorbed by blood at faster rate compared to the attached fraction. The unattached fraction is predominantly responsible for radiation dose received by the target cells in the bronchial epithelium. Thus, it is important to measure both the size modes for accurate dose assessment. The alpha decays imparting the radiation dose of greatest significance are from $^{219}$Po and $^{214}$Po in $^{222}$Rn series, $^{212}$Bi and $^{212}$Po are for $^{220}$Rn series. The dose to the bronchial basal cells from unattached $^{218}$Po is up to 38 times higher than that from attached radon progeny because of its efficient deposition in the upper bronchial tree\textsuperscript{10}.

The attached and unattached decay product concentration in the indoor atmosphere is a crucial parameter in dose assessment. Chamberlain and Dyson demonstrated the preferential deposition of unattached radon progeny in the human upper respiratory tract\textsuperscript{11}. In this study the attached and unattached fractions of $^{222}$Rn and $^{220}$Rn progeny concentrations have been determined using deposition based radon and thoron progeny sensors (DRPS/DTPS)\textsuperscript{12-15} (Figure 1). The study was carried out in the Chavara and Neendakara hamlets of Kollam district, in Kerala where the high background radiation exists.

**Experimental**

In order to assess the equilibrium equivalent decay product concentrations, direct radon / thoron progeny sensors (DRPS/DTPS) were deployed in 38 dwellings of Chavara and Neendakara hamlets of Kollam district, Kerala, which is a high background radiation area (HBRA). The houses selected for the study were at least 10 years old. The codes given are C and N respectively for Chavara and Neendakara. The study area is shown in figure 3. The ventilation conditions of the dwellings were noted in terms of the number of windows and doors of the room and the period of their usage. The dosimeters were suspended 40 cm away from the walls and 1.7 meters above the floor. The exposure time was 100 days. The retrieved dosimeter films (LR-115 Type II) were etched using 2.5N NaOH solution at 60\degree C for 60 minutes without stirring and scanned by spark counter. The single entry pin hole based dosimeters were deployed along with DRPS/DTPS for the measurement of $^{222}$Rn and $^{220}$Rn. The measurement protocols and dimensions of the dosimeter have been discussed elsewhere\textsuperscript{16}.

Absorber mounted LR115 type II detectors are used in DTPS and DRPS. The sensors selectively detect $\alpha$-particles from $^{214}$Po and $^{212}$Po. The absorber in DTPS is 50\,µm Al mylar, which permit only the $\alpha$-particles of energy 8.78\,MeV emitted from $^{212}$Po to pass through it. In DRPS the absorber has an effective thickness of 37\,µm. It is a combination of 25\,µm Al mylar and 12\,µm cellulose nitrate peeled off from LR115. DRPS selectively detects 7.69\,MeV $\alpha$-particles emitted from $^{214}$Po. The equilibrium equivalent decay product concentrations were estimated from the track densities using the sensitivity factors discussed by Mishra and Mayya\textsuperscript{12}. Since DTSP is not sensitive to radon progeny, its track density can be directly used to calculate EETC. But in DRPS, both and radon and thoron progenies register tracks. So it is necessary to subtract the track density of thoron progeny estimated from the DTSPS using the equation (1)

\[
\text{Track density}_{\text{only due to Rn progeny}} = \text{Total track density}_{\text{DRPS}} - \frac{\eta_{\text{TT}}}{\eta_{\text{TT}}} \text{Track density}_{\text{DTPS}}
\]

(1)

\[
\eta_{\text{TT}} = 0.083, \text{ track registration efficiency of } \text{thoron progeny in DRPS}
\]

\[
\eta_{\text{RT}} = 0.01, \text{ track registration efficiency of } \text{thoron in DTSP}
\]

The equilibrium equivalent $^{222}$Rn and $^{220}$Rn concentrations were calculated using the formulae

\[
\text{EETC}_{\text{A+U}} = \frac{\text{Track density}_{\text{DTPS}}}{K_T \times \text{Exposure period (d)}}
\]

(2)

\[
\text{EERC}_{\text{A+U}} = \frac{\text{Track density}_{\text{Only due to Rn progeny}}}{K_R \times \text{Exposure period (d)}}
\]

(3)

where $K_T$ and $K_R$ are calibration factors for DTSP and DRPS, respectively. The sensitivity factors for DTSP and DRPS in indoor atmosphere have been determined by Mishra et al.\textsuperscript{12} as 0.94 \, Tracks \, cm\textsuperscript{-2} \, d\textsuperscript{-1}/EETC (Bq m\textsuperscript{-3}) for DTSP and 0.09 \, Tracks \, cm\textsuperscript{-2} \, d\textsuperscript{-1}/EERC (Bq m\textsuperscript{-3}) for DRPS. The minimum detection limit of DTSP and DRPS were 0.1 Bq m\textsuperscript{-3} and 1 Bq m\textsuperscript{-3} respectively\textsuperscript{14}.

The attached progeny concentration was obtained by wire mesh capped DTSP/DRPS. It consists of DTSP/DRPS capped with a 200 type wire screen as shown in figure 2. The tracks recorded are converted to attached decay product concentrations (EECA / EECTA) using the equations (4) and (5)\textsuperscript{12,13}.

\[
\text{EECT}_A = \frac{\text{Track density}_{\text{Capped DTSP}}}{K_T \times \text{Exposure period (d)}}
\]

(4)

\[
\text{EERC}_A = \frac{\text{Track density}_{\text{Capped DRPS}} - \text{EECT}_A}{K_R \times \text{Exposure period (d)}}
\]

(5)
Where the $K_{TT}$ and $K_{RR}$ are the calibration factors which have values 0.33 $\text{Tr cm}^{-2} \text{d}^{-1}/\text{EETC (Bq m}^{-3})$ and 0.04 $\text{Tr cm}^{-2} \text{d}^{-1}/\text{EERC (Bq m}^{-3})$. The unattached progeny concentration was obtained by subtracting attached progeny concentration from total (attached + unattached) decay product concentration. The annual indoor inhalation dose for $^{222}\text{Rn}$ and $^{220}\text{Rn}$ were determined using the dose conversion factors suggested by UNSCEAR-2008\textsuperscript{17}.

An attempt was made to investigate the effect of ventilation based on the design of houses on radon, thoron and progeny levels. For this purpose, the dwellings were classified into 3 different categories, namely: poorly ventilated, partially ventilated and well ventilated. The categorization was made based on the period of use and number of purposed-built openings namely windows and doors. All dwellings we studied had only natural ventilation. Rooms with small dimensions having one door and one small window or without window not or rarely opened were treated as poorly ventilated. Well ventilated dwellings had two or more doors and windows kept opened at least 8 hours per day. The houses we studied were made of cement, stones and bricks. The roofs are made with cement concrete and the floors were made with cement and concrete.

Results and Discussion

The attached and unattached progeny concentrations of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ are presented in table 1. Total progeny concentration (attached + unattached) concentration of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ in the Chavara region was found to vary from 8.8±1 to 22.5±2 Bq m\(^{-3}\) and 1.1±1.0 to 3.3±0.2 Bq m\(^{-3}\) respectively. For Neendakara region total progeny concentrations were found to vary from 11.1±1 to 22.2±2 Bq m\(^{-3}\) and 1.1±0.1 to 4.0±0.2 Bq m\(^{-3}\) respectively. In general total progeny concentrations were found to be higher in poorly ventilated houses due their accumulation. The EERC\(_A\) and EERC\(_U\) in the dwellings of the study area ranged from 8±2.8 to 21±4.5 Bq m\(^{-3}\) and 0.5±0.1 to 2.4±1.4 Bq m\(^{-3}\), respectively. The EETCA and EETCU in the dwellings vary from 1.0±0.9 to 3.8±1.9 Bq m\(^{-3}\) and 0.1±0.1 to 0.2±0.1 Bq m\(^{-3}\), respectively. In all dwellings, the unattached fractions of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ have been found to be quite low. This can be accounted for the high humidity (up to 80\%) and high aerosol concentration of the coastal region. The levels of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ in the regions were found to be 43.6±11.1 Bq m\(^{-3}\) and 57.1±19.2 Bq m\(^{-3}\) respectively, the maximum being 75±8.6 and 95±9.7 Bq m\(^{-3}\), respectively.

The average values for thoron levels in the study area were found to be higher than the global average of 10 Bq/m\(^3\) as well as the national average of 12.2 Bq/m\(^3\) owing to the thorium rich monazite sand in the regions.

The annual effective dose for $^{222}\text{Rn}$ and $^{220}\text{Rn}$ in the study area was found to vary from 1.48±0.9 to 3.7±1.6 mSv y\(^{-1}\) as seen in table 1. In many locations the annual effective dose estimate was found to be more than the recommended level of 3 mSv y\(^{-1}\). The unattached fraction of $^{222}\text{Rn}$ progeny (UF\(_{\text{Rn}}\)) in the area varies from 0.04 to 0.15 and it was found to be higher in well ventilated houses. The unattached fraction of $^{220}\text{Rn}$ progeny (UF\(_{\text{Th}}\)) ranged from 0.03 to 0.09 with an average value of 0.069. It is noticed that there exists no relationship between UF\(_{\text{Th}}\) with ventilation conditions of the dwellings as seen from the figure 7.

![Figure 1. DRPS/DTPS Progeny sensors](image1)

![Figure 2. Wiremesh DRPS/DTPS Progeny sensors](image2)
Figure 3. Study area

Table 1. The unattached fractions of $^{222}$Rn and $^{220}$Rn progeny in Chavara and Neendakara hamlets of Kollam district

| CODE | EETC$_{A+U}$ (Bqm$^{-3}$) | EETC$_{A}$ (Bqm$^{-3}$) | EETC$_{U}$ (Bqm$^{-3}$) | UF$_{A}$ | AEDT (mSv a$^{-1}$) | EERC$_{A+U}$ (Bqm$^{-3}$) | EERC$_{A}$ (Bqm$^{-3}$) | EERC$_{U}$ (Bqm$^{-3}$) | UF$_{Rn}$ (mSv y$^{-1}$) | Total dose (mSv y$^{-1}$) |
|------|---------------------------|------------------------|------------------------|----------|---------------------|------------------------|------------------------|------------------------|---------------------|----------------------|
| N1   | 2.20±0.2 2.00±0.2          | 0.20                   | 0.09                   | 1.29     | 12.5±1              | 11.5±1                 | 1.0                    | 0.08                   | 1.10                | 2.39                  |
| N2   | 1.56±0.1 1.40±0.1          | 0.10                   | 0.09                   | 1.20     | 14.3±1              | 13.3±1                 | 1.0                    | 0.07                   | 1.26                | 2.46                  |
| N3   | 3.33±0.2 3.13±0.2          | 0.20                   | 0.06                   | 1.93     | 12.0±1              | 11.4±1                 | 0.6                    | 0.05                   | 0.79                | 2.72                  |
| N4   | 1.86±0.1 1.73±0.1          | 0.13                   | 0.07                   | 1.72     | 15.0±2              | 14.1±1                 | 0.9                    | 0.06                   | 1.32                | 3.04                  |
| N5   | 4.00±0.2 3.80±0.2          | 0.20                   | 0.05                   | 1.86     | 14.3±1              | 13.3±1                 | 1.0                    | 0.07                   | 1.26                | 3.12                  |
| N6   | 2.22±0.2 2.02±0.2          | 0.20                   | 0.09                   | 1.29     | 12.5±1              | 12.0±1                 | 0.5                    | 0.04                   | 1.65                | 2.94                  |
| N7   | 2.50±0.2 2.35±0.2          | 0.15                   | 0.06                   | 1.93     | 16.0±2              | 15.2±1                 | 0.8                    | 0.05                   | 1.41                | 3.34                  |
| N8   | 1.56±0.1 1.42±0.1          | 0.10                   | 0.04                   | 1.93     | 15.0±1              | 14.4±1                 | 0.6                    | 0.04                   | 1.32                | 3.25                  |
| N9   | 1.86±0.2 1.73±0.1          | 0.10                   | 0.09                   | 1.20     | 14.3±1              | 13.3±1                 | 1.0                    | 0.07                   | 1.26                | 3.12                  |
| N10  | 3.33±0.2 3.13±0.2          | 0.20                   | 0.06                   | 1.93     | 12.0±1              | 11.4±1                 | 0.6                    | 0.05                   | 0.79                | 2.72                  |
| N11  | 1.86±0.1 1.73±0.1          | 0.13                   | 0.07                   | 1.72     | 15.0±2              | 14.1±1                 | 0.9                    | 0.06                   | 1.32                | 3.04                  |
| N12  | 4.00±0.2 3.80±0.2          | 0.20                   | 0.05                   | 1.86     | 14.3±1              | 13.3±1                 | 1.0                    | 0.07                   | 1.26                | 3.12                  |
| N13  | 2.22±0.2 2.02±0.2          | 0.20                   | 0.09                   | 1.29     | 12.5±1              | 12.0±1                 | 0.5                    | 0.04                   | 1.65                | 2.94                  |
| N14  | 2.50±0.2 2.35±0.2          | 0.15                   | 0.06                   | 1.93     | 16.0±2              | 15.2±1                 | 0.8                    | 0.05                   | 1.41                | 3.34                  |
| N15  | 1.56±0.1 1.42±0.1          | 0.14                   | 0.09                   | 1.20     | 15.0±2              | 14.1±1                 | 0.9                    | 0.06                   | 1.07                | 2.27                  |
| N16  | 1.86±0.2 1.73±0.1          | 0.13                   | 0.07                   | 1.23     | 11.1±1              | 10.1±1                 | 1.0                    | 0.09                   | 0.65                | 1.88                  |
| N17  | 2.83±0.2 2.66±0.2          | 0.17                   | 0.06                   | 1.46     | 11.1±1              | 10.1±1                 | 1.0                    | 0.09                   | 0.64                | 2.10                  |
| N18  | 2.00±0.1 1.90±0.1          | 0.10                   | 0.05                   | 1.16     | 12.9±1              | 12.0±1                 | 0.9                    | 0.07                   | 0.85                | 2.01                  |
| N19  | 1.43±0.1 1.33±0.1          | 0.10                   | 0.07                   | 1.10     | 13.8±1              | 12.7±1                 | 1.1                    | 0.08                   | 1.21                | 2.31                  |
| N20  | 1.11±0.1 1.01±0.1          | 0.10                   | 0.09                   | 0.74     | 14.3±1              | 13.3±1                 | 1.0                    | 0.07                   | 1.11                | 1.85                  |
| N21  | 1.43±0.1 1.33±0.1          | 0.10                   | 0.07                   | 1.10     | 13.3±1              | 12.1±1                 | 1.2                    | 0.09                   | 0.93                | 2.03                  |
| N22  | 1.56±0.1 1.42±0.1          | 0.14                   | 0.09                   | 0.90     | 8.8±1               | 8.0±1                  | 0.8                    | 0.09                   | 0.83                | 1.73                  |
| N23  | 2.50±0.2 2.40±0.1          | 0.10                   | 0.04                   | 1.93     | 11.3±1              | 10.4±1                 | 0.9                    | 0.08                   | 0.93                | 2.86                  |
| N24  | 1.67±0.1 1.57±0.1          | 0.10                   | 0.06                   | 1.55     | 11.0±1              | 9.9±1                  | 1.1                    | 0.10                   | 0.65                | 2.20                  |
| N25  | 1.71±0.2 1.59±0.1          | 0.12                   | 0.07                   | 0.99     | 14.6±1              | 13.6±1                 | 1.0                    | 0.07                   | 0.84                | 1.83                  |
| N26  | 3.33±0.2 3.23±0.2          | 0.10                   | 0.03                   | 1.93     | 13.8±1              | 12.7±1                 | 1.1                    | 0.08                   | 1.01                | 2.94                  |
| N27  | 2.14±0.1 1.99±0.1          | 0.15                   | 0.07                   | 1.66     | 16.0±1              | 15.0±1                 | 1.0                    | 0.06                   | 1.24                | 2.90                  |

Note: Poorly ventilated

Partially ventilated
| Location | Gas Concentration ($Bq m^{-3}$) | Ventilation Rate | EERC$_A$ | EERC$_U$ | AEDR ($mSv a^{-1}$) | AEDT ($mSv a^{-1}$) |
|----------|-------------------------------|-----------------|----------|----------|--------------------|--------------------|
| N14      | 4.00±0.2 3.80±0.2             | 0.20            | 0.05     | 1.86     | 16.0±1             | 14.4±1             |
| N15      | 1.10±0.1 1.00±0.1             | 0.10            | 0.09     | 0.57     | 10.0±1             | 8.9±1              |
| N16      | 2.00±0.1 1.90±0.1             | 0.10            | 0.05     | 0.77     | 15.0±1             | 12.9±1             |
| N17      | 1.43±0.1 1.33±0.1             | 0.10            | 0.07     | 0.66     | 18.2±1             | 16.2±1             |
| N18      | 1.33±0.1 1.21±0.1             | 0.12            | 0.09     | 0.88     | 13.1±1             | 11.1±1             |
| N19      | 2.00±0.1 1.88±0.1             | 0.12            | 0.06     | 1.16     | 20.0±2             | 18.0±2             |
| N20      | 1.43±0.1 1.33±0.1             | 0.10            | 0.07     | 0.66     | 16.2±1             | 14.1±1             |
| N21      | 2.00±0.1 1.88±0.1             | 0.12            | 0.06     | 1.55     | 22.2±2             | 20.2±2             |
| C12      | 2.86±0.2 2.66±0.2             | 0.20            | 0.07     | 2.21     | 17.7±1             | 15.4±1             |
| C13      | 1.10±0.1 1.00±0.1             | 0.10            | 0.09     | 0.86     | 14.0±1             | 11.9±1             |
| C14      | 1.43±0.1 1.33±0.1             | 0.10            | 0.07     | 0.83     | 14.9±1             | 13.3±1             |
| C15      | 1.10±0.1 1.00±0.1             | 0.10            | 0.09     | 1.03     | 15.8±1             | 13.9±1             |
| C16      | 1.11±0.1 1.01±0.1             | 0.1            | 0.09     | 0.89     | 16.2±1             | 14.1±1             |
| C17      | 2.60±0.2 2.47±0.2             | 0.13            | 0.05     | 1.51     | 16.0±1             | 13.6±1             |

Figure 4. Distribution of $^{222}$Rn and $^{220}$Rn in the study area.

Figure 5. The variation of EERC$_A$ and EERC$_U$ with ventilation rate.

Figure 6. The variation of annual effective dose due to radon and thoron (AEDR & AEDT) in the dwellings of Chavara and Neendakara.
Conclusion

The EEC of $^{222}$Rn and $^{220}$Rn progeny is higher in poorly ventilated rooms. The attached progeny concentration of $^{222}$Rn and $^{220}$Rn is found to be greater than the unattached progeny concentration. The unattached progeny concentration of $^{222}$Rn was found to be more in well ventilated rooms. This is expected for well ventilated rooms as the aerosol concentration would be less in such rooms. The levels of unattached fraction of radon decay products obtained in the present study is in good agreement with values reported by R Mehra et al. $^{19}$ $^{220}$Rn unattached progeny has not shown any relationship with ventilation rate. The average annual effective dose in the study area is $2.48\pm 0.57$ mSv$^{-1}$. A more comprehensive and prolonged study is necessary to understand the seasonal variations and influence of other confounding factors on dose estimate.

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