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Indoor radon, thoron and their progeny levels

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ABSTRACT

Among all possible sources of radiation dose to human beings, inhalation of radon and its progeny contributes a lot to the dose. High concentrations of radon and its daughter products in mines have led to an increase in the incidence of lung cancer among mine workers. There is correlation of lung cancer risk for the people working in mines. For the long term measurements more than 100 dwellings were selected for the measure of radon, thoron and their progeny concentrations using solid state nuclear track detectors. By knowing the track density of films through spark counter, and sensitivity factor for bare, filter and membrane; the concentrations of radon, thoron and their progeny were calculated. The radon, thoron and their progeny levels were found to vary from wall to wall. The variation may be attributed due to random distribution of radioactive rock used in the construction of buildings. The radon, thoron and their progeny levels in different types of floorings, wall, room and building material are well within the admissible limit. © 2009 Trade Science Inc. - INDIA

KEYWORDS

Radon;
Thoron;
Walls;
Floors;
Rooms;
Building materials;
SSNTD;
Dwellings.

1. INTRODUCTION

The origin of radon and thoron in the Earth's crust stems directly from the uranium, thorium, and their decay products distributed in minute quantities in the ground within few metres of the Earth's surface^[1-5]. Radon atoms generated in Earth's crust enter the pore spaces and are then transported by diffusion and advection through this space until they decay or are released into the atmosphere. The amount of radon that escapes depends on the amounts of ²²⁶Ra and ²³²Th in the ground, the type of the soil cover, porosity, damp-

ness and temperature of the soil. Once the gas is formed it migrates to a significant distance from the site of generation, even during their brief half-lives. Radon properties have led to its use as a geographical tracer for locating buried faults and geological structures, in exploring the uranium from mines, and for predicting the earthquakes^[6]. Among the isotopes of radon, the greatest attention is given to ²²²Rn as it has the longest half-life. In addition, the short-lived daughters, polonium, lead and bismuth are valuable since they are natural tracers in the atmosphere^[7].

In addition, there is an increasing interest in the sub-

ject of indoor radioactivity particularly to the health effects that might result from prolonged exposure to airborne radio nuclides. Most of the literatures have focused on radon and its progeny ^{218}Po , ^{214}Pb , ^{214}Bi since they are known to make a major contribution to dose from natural radioactivity^[8-10]. Radon and its daughters are the most important radionuclide present in the ambient air as well as in indoor atmosphere. Radon being a gas escapes from the soil by diffusion and mixes with the air by horizontal and vertical transportation. The radon daughters attach to submicron sized aerosols particles and causes hazards to lungs during inhalation^[11,12].

The concentration of radon builds up inside the houses due to exhalation from soil and walls. Radon exhalation rate from building materials mainly depends on the physical conditions of materials, radium content, porosity of the material used in the floor, walls and ceilings of house. Exhalation rates are also influenced by the meteorological parameters such as temperature, humidity, rainfall, etc. In the present study the radon/thoron concentration in different types of flooring, walls and building material are determined.

2. MATERIALS AND METHOD

2.1. Solid state nuclear track detectors

The concentrations of radon, thoron and their progeny are measured in dwellings of some parts of Mysore city using Solid State Nuclear Track Detectors (SSNTD), which are thin sheets of dielectric materials such as cellulose nitrate (CN) and polycarbonate. SSNTD's are more sensitive to alpha compared to beta and gamma radiations. The films are less influenced by the moderate humidity, heat and light. For indoor measurements normally LR-115 TYPE II (Kodak Pathe, France) plastic track detectors are preferred.

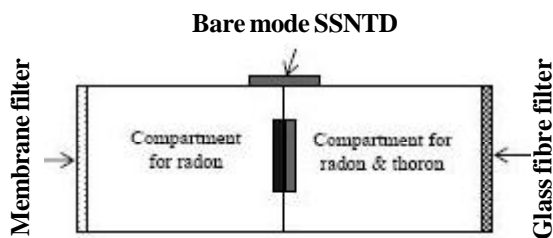


Figure 1 : The double chamber dosimeter cup

The double chamber dosimeter cup used for monitoring radon, thoron and their progeny is shown in figure 1. The chamber has a length of 45 mm and a radius of 31mm. The films used in the dosimeter are of approximately 12 μm thickness. The SSNTD placed in left compartment that measures only radon, which diffuses into it from the ambient air through a semi-permeable membrane. These membranes have permeability constants in the range of $10^{-12} - 10^{-11} \text{ m}^2 \text{ s}^{-1}$ and allow more than 95% of the radon gas to diffuse and reduce thoron concentration to the extent of 1% or even less^[13]. On the right side, the glass fiber filter paper placed in the compartment allows both radon and thoron gas to diffuse in and hence the tracks on SSNTD are measure of concentration of both the gases. The SSNTD exposed in the bare mode, placed on the top surface of the dosimeter registers alpha tracks attributable to the air borne concentrations of both the gases and their progeny^[14]. These dosimeters are suspended from the midpoint of the house at a height of 2 m from the ground level. At the end of the stipulated period of exposure, usually 3 months, the dosimeters are retrieved and all the three SSNTD's are etched with 10% sodium hydroxide solution for one hour at 60 $^{\circ}\text{C}$ ^[15]. The track density of alphas in the film is determined using a spark counter. This exposure cycle has been extended in a time integrated four quarterly cycles to cover all the four seasons of a calendar year to evaluate the annual radon/thoron and their progeny levels.

The radon/thoron levels and their progeny working level concentrations are calculated by the following relations^[14].

$$C_R (\text{Bq m}^{-3}) = T_m / (d S_m)$$

$$C_T (\text{Bqm}^{-3}) = (T_f - d C_R S_{rf}) / (d S_{rf})$$

where T_m is the track density of the film in membrane compartment, d is the period of exposure in days, S_m refers to the sensitivity factor of membrane compartment, T_f is the track density of the film in filter compartment, S_{rf} is the Sensitivity of ^{222}Rn in filter compartment, C_R and C_T is the concentration of ^{222}Rn and ^{220}Rn , respectively.

$$R_n (\text{m WL}) = (C_R F_R) / 3.7$$

$$R_T (\text{m WL}) = (C_T F_T) / 0.275$$

where R_n and R_T refers to the progeny concentrations of ^{222}Rn and ^{220}Rn , respectively.

$$F_R = 0.104 f_{RA} + 0.518 f_{RB} + 0.37 f_{RC}$$

$$F_T = 0.91 f_{TB} + 0.09 f_{TC}$$

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where f_{RA} , f_{RB} and f_{RC} are the activity fractions with respect to parent gas.

But, F_R and F_T represents the equilibrium factors for ^{222}Rn and ^{220}Rn progeny corresponding to the extracted ventilation rate^[14]. Equilibrium factor is determined using the working level concentrations, and the inhalation dose (mSvy^{-1}) is estimated by using UNSCEAR^[16]:

$$D = 0.007 \times [(0.17 + 9F_R) C_R + (0.11 + 32F_T) C_T]$$

2.2. Estimation of activity of radium

The activity of ^{226}Ra in the soil and building materials is estimated by using the gamma ray spectrometer. The soil samples were collected at several locations near the houses of different types of construction in Mysore city. The places that are free from surface runoff during heavy rain were carefully selected. An area of about 0.5 m^2 was chosen and the vegetation and roots were removed, if any. Then about 2 kg of soil samples were collected from these places at a depth of approximately 20cm from the earth's surface^[17]. After the collection, the samples were mixed thoroughly and extraneous materials such as plants, debris, big pieces of stones, pebbles etc. were removed. Composite samples of about 2 kg was taken and sealed in a polythene bag. These samples were dried in oven maintained at a temperature of 110°C for about 10-12 hours. Then the samples were powdered and sieved through 150 micron sieves, weighed and sealed in a 300 ml plastic container, and kept for a month before counting for gamma spectrometry, in order to ensure that radioactive equilibrium was reached between ^{226}Ra , ^{222}Rn and its progeny. The spectrometer consisted of n-type HPGe coaxial detector of 17% efficiency having a resolution of 1.75 keV at 1.33 MeV and 641 eV at 5.9 keV and coupled to a 4096 channel MCA. The detector is housed in a lead shield to cease the background radiation entering from outside. The gamma spectrometric procedure is followed to estimate the activity of ^{226}Ra . The gamma peak of energy 609.51 keV with intensity of 46.1 % is used as proxy for the quantitative determination of ^{226}Ra by gamma spectrometry. The activity of radionuclide is calculated using the relation:

$$\text{Activity (Bq kg}^{-1}\text{)} = (S \pm \sigma) 10^7 / (E W A)$$

where S is the net counts/s under the photo peak of interest, σ is the standard deviation of S, E is the counting efficiency(%),

A is the gamma abundance (%) for the radionuclide, W is the mass of the sample (g)

3. RESULTS AND DISCUSSION

Several measurements are carried out using SSNTDs for the concentrations of radon/thoron in dwellings. The variation of the concentration is mainly because of ventilation rate in houses and the materials used. Sathish et al.^[18] have measured the concentration of radon, thoron and their progeny in different types of buildings at various locations in Mysore city. They have reported the arithmetic mean values of concentrations of radon and thoron in indoor atmosphere ranged from 9.20 to 58.02 with a median of 34 Bq m^{-3} and 7.21 to 59.27 with a median of 33 Bq m^{-3} , respectively. The arithmetic mean of progeny concentrations varied from 0.033 to 2.52 with a median of 1.29 and 0.074 to 47.04 with a median of 20.7 mWL, respectively. The reported dose to the selected population of Mysore city was 1.6 m Sv y^{-1} .

3.1 Different floorings

Radon, thoron and their progeny levels were measured in different types of floorings of houses at different locations in Mysore city. The measurements were made covering all the four seasons in a calendar year and the integrated data is shown in figure 2.

The walls and flooring of the houses are of different materials^[19]. The materials used for construction of buildings are sufficiently porous and allow ^{222}Rn to en-

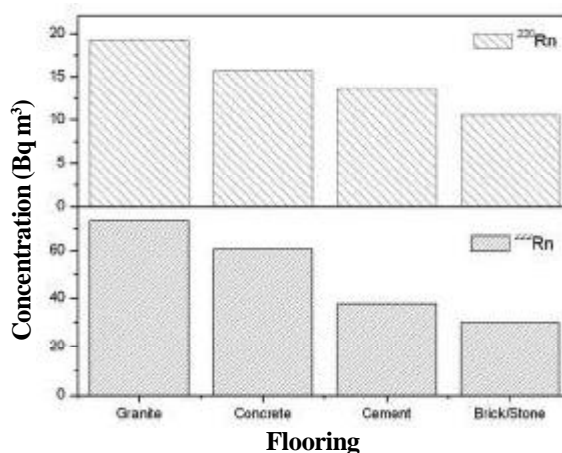


Figure 2 : Variation of ^{222}Rn and ^{220}Rn in different floorings

TABLE 1: Radium thorium in different building materials^[20]

Building materials	Radium(Bq kg ⁻¹)	Thorium(Bq kg ⁻¹)
Brick	12.5	25
Cement	25.6	22
Concrete	26.8	24
Sand	5.4	43
Granite	54.5	46

TABLE 2: Radon exhalation in different materials

Types of surface	²²² Rn exhalation rate (m. Bq m ⁻² .S ⁻¹)
Ground floor	
Cement flooring red oxide coated	3.29±0.42
Concrete surface	2.77±0.31
Heavy concrete surface	0.95±0.14
Mosaic flooring	1.71±0.46
Bare (Huts)	4.98±0.89
Soil/Brick	5.49±1.00

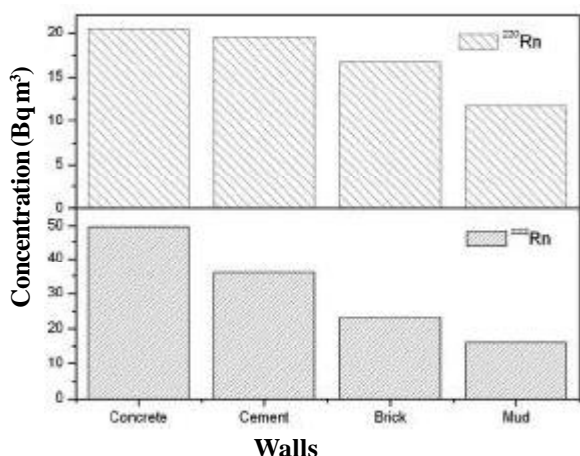


Figure 3 : Variation of ²²²Rn and ²²⁰Rn in different walls

ter into the indoor atmosphere. The figure 2 also shows that the high ²²²Rn concentration is observed in loose cement flooring houses and low concentration in stone flooring houses. Loose cement flooring houses show high ²²²Rn exhalation rate than stone flooring houses. This is due to low porosity and low diffusion coefficient in the stone flooring than in ordinary cement flooring houses. The data shows that in the houses with stone flooring, concentrations of ²²²Rn is less but in the house with granite flooring it is slightly higher because granite contains high concentrations of ²²⁶Ra. This can be clearly seen in TABLE 1. The geometric mean values of radium and thorium are 19.07 and 30.04 Bq kg⁻¹. This feature for Mysore environment is reported by Sannappa et al.,^[20] and shown that there is a positive correlation between the exhalation rate and concentration in building materials. Granite samples show higher

²²²Rn exhalation rate than marble, and the correlation of the ²²⁶Ra content of granite with ²²²Rn exhalation and ²²²Rn concentration is observed by several researchers^[21-24].

Mosaic flooring houses show slightly less ²²²Rn exhalation rate than cement flooring houses. The huts have mud walls, bare flooring and poor ventilation. The ²²²Rn concentration in huts is lower than in concrete and mosaic flooring houses even though ²²²Rn exhalation from bare flooring is higher than other types of flooring. This may be attributed to the possibility of out flow of air from huts through the coconut dry leaves that form the roof. Further the fume emitting from cooking stove would increase the air pressure inside dwellings and forcing it outside. ²²²Rn exhalation in mud walls is less compared to the cemented walls. High ²²²Rn exhalation from concrete walls may be responsible for increasing the concentration in cement and mosaic flooring houses. This may be due to the high ²²²Rn concentration in the concrete floorings and also because of the porous floor concrete slab through which ²²²Rn could easily enter the houses. The exhalation from brick wall with cement plastering is also high as shown in TABLE 2. In addition to this the high indoor ²²²Rn concentration could be attributed to cracks in the floor as a consequence of poor quality of materials used for construction^[25].

3.2. Different walls

Annual average concentration of ²²²Rn, ²²⁰Rn and their progeny levels for different types of wall are shown in figure 3. The ²²²Rn, ²²⁰Rn and their progeny levels are found to vary from wall to wall. The variation may be due to random distribution of radioactive rock species used ignorantly in the construction of the houses^[26,27]. The reported average values of porosity of the soil and building materials are 0.25 and 0.15^[28] respectively. The ²²²Rn exhalation rate is higher in soil than in concrete, whereas building may contain several tons of concrete. The contribution of this to indoor ²²²Rn is relatively low because of low escape rate of ²²⁶Ra. ²²²Rn exhalation rate also depends on the thickness of wall as evident in TABLE 2. UNSCEAR^[16] reports the exhalation rates from walls and floor of half slab thickness 0.1m – 0.05m as 1.6 mBqm⁻²s⁻¹ and 0.8 mBqm⁻²s⁻¹, respectively. Ramachandran et al.^[29] have

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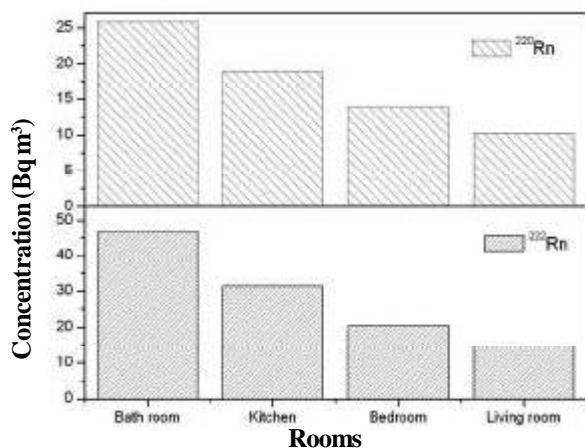


Figure 4 : Variation of ²²²Rn and ²²⁰Rn in different rooms

quantified these results and reported the exhalation rate of ²²²Rn from building materials. The reported values for concrete and brick vary from $0.43-1.3 \times 10^{-5}$ and 0.1×10^{-5} Bqkg⁻¹s⁻¹ for USA, 0.47×10^{-5} 0.017×10^{-5} Bqkg⁻¹s⁻¹ for Denmark and 0.32 and 0.16×10^{-5} Bq kg⁻¹s⁻¹ for USSR. The reported values for Hungary are 0.78×10^{-5} and 0.39×10^{-5} Bqkg⁻¹s⁻¹, respectively. Vinaya Kumar et al.^[30] have seen that the radon/thoron levels are more in mud walls compared to other type of walls. They also reported that the concentration levels in dwellings of mud plastered walls can be expected more and is generally taken from local soil for constructions.

3.3. Different rooms

The figure 4 shows the annual average concentrations of ²²²Rn in different rooms of the same house. One can clearly see in the figure that there is high concentration in bathroom compared to the other rooms in the houses. The differences were clearly present, bathroom was found to have higher ²²²Rn concentration, kitchen ranked second, next bed rooms and living room the least. Bed rooms might be expected to be least ventilated, on the average based upon limited use patterns and bath rooms may receive some additional ²²²Rn due to ²²²Rn dissolved in water^[31]. ²²²Rn is shown to be released in spray from faucets or shower fixture^[32,33]. Air in living rooms on the other hand is most readily diluted due to outdoor air blow. This can be correlated with concentrations of ²²²Rn in water^[20] and the activity of ²²⁶Ra.

Yadagiri reddy et al.^[34] have reported that the ra-

don concentration varied from 17 to 311 Bq m⁻³ and progeny concentration varied between 0.1 to 20mWL with an average of 62 ± 45 Bq m⁻³ and 4.7 ± 4.1 mWL, respectively. In addition they have observed that bed rooms have relatively higher concentrations of radon, thoron followed by kitchen, front room and hall. This may be due to less atmospheric pressure and less ventilation in the bed room.

4. SUMMARY

The concentration level of ²²²Rn and ²²⁰Rn in dwellings depend on various factors such as soil beneath, local geology, the house construction materials, micro-climatic parameters and last but not the least life style in the dwelling since higher concentration in the poor ventilated rooms (bed room, kitchen, bath room) have been observed. Therefore it is suggested that the residential rooms must be well ventilated and free from ²²²Rn rich materials to reduce the health hazards due to ²²²Rn. Indoor ²²²Rn and ²²⁰Rn concentration depends also on ²²²Rn exhalation rate of the flooring and ventilation condition. Higher concentrations of both ²²²Rn and ²²⁰Rn have been observed where the exhalation rate is more. The activity of ²²⁶Ra in the soils of Mysore region is lower than the global average value and indoor ²²²Rn and ²²⁰Rn concentrations are comparable with the global average.

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