

Concentration of radon, thoron and their progeny levels in different types of floorings, walls, rooms and building materials

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Background: Radon, thoron and their progenies are the most important contributions to human exposure from natural sources. Radon exists in soil gas, building materials, Indoor atmosphere etc. Among all the natural sources of radiation dose to human beings, inhalation of radon contributes a lot. The work presented here emphasizes the long term measurements of radon, thoron and their progeny concentrations in about 100 dwellings using solid state nuclear track detectors. **Materials and Methods:** Measurements were made using dosimeters and the concentrations were estimated by knowing the track density of films through spark counter, and sensitivity factor for bare, filter and membrane films. **Results:** Presence of radon and thoron in houses is the effect of several aspects such as the activity concentrations of uranium, radium and thorium in the local soil, building materials, ventilation of houses and also entry of radon into houses through the cracks in floor/wall. **Conclusion:** The observations reveal that the concentrations of radon and/or thoron are relatively higher in granite than in concrete, cement and bricks. In continuation to this the concentration observed in bathrooms is more compared to kitchen, bedroom and living rooms. This study discloses that the residential rooms of good ventilation will avoid the health hazards due to radon and its rich materials. *Iran. J. Radiat. Res., 2009; 7 (1): 1-9*

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INTRODUCTION

The origin of radon and thoron in the atmosphere is due to the presence of uranium and thorium in the rocks and soil of the Earth's surface (1-3). 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 and/or released into the atmosphere.

The amount of radon that escapes depends on the amounts of ²²⁶Ra and ²³²Th in the ground, type of the soil cover, porosity, dampness, temperature and moisture of the soil (4). In addition, there are several mechanisms to contribute for the escape of radon from its origin to the atmosphere such as alpha-recoil, emanation and so on. Once the gas is formed it migrates to a significant distance from the site of generation, even during their brief half-lives (5). 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. The short-lived daughters, ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi and ²¹⁴Po are valuable since they are natural tracers in the atmosphere (7).

Now a days, there is an increasing interest in the subject 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 ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi since they are known to make a major contribution to dose from natural radioactivity (8, 9). 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

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the soil by diffusion and mixes with the air by horizontal and vertical transportation. The radon daughters attach to submicron sized aerosol particles in the atmosphere and will expose the lungs to radiation when inhaled^(10, 11). However, there are only limited studies related to the measurements of thoron in the environment^(12, 13) since it is assumed that the inhalation dose to the general population from thoron and its progeny is only about 10% of the inhalation dose due to radon⁽¹⁴⁾. In general, this type of studies are important in two ways, viz., any radiological impact assessment of nuclear facilities, either existing or those to be set up in the future, requires information on the exposure due to natural radiation prevalent in their vicinity. And the radiation risk coefficients are fairly well established at high dose rates, whereas little is known about the effects of radiation at low dose rates⁽¹⁵⁾.

The concentration of radon in houses builds up due to the exhalation from soil and walls. Entry of radon into the houses mainly depends on building materials and on the physical conditions of materials, radium content, porosity of the material used in the floor, walls and ceiling of the houses. The entry of gas is also influenced by the meteorological parameters such as temperature, humidity, rainfall, etc. The radon, thoron and their progeny concentration in different types of flooring, walls, rooms and building materials are determined and their variations are discussed in detail.

MATERIALS AND METHODS

Solid State Nuclear Track Detectors

The concentrations of radon, thoron and their progenies are measured in the dwellings of some parts of Mysore city using Solid State Nuclear Track Detectors (SSNTDs), which are thin sheets of dielectric materials such as cellulose nitrate and polycarbonate. SSNTD's are more sensitive to alpha compared to beta and gamma

radiations. For Indoor measurements LR-115 TYPE II (Kodak Pathe, France) plastic track detectors were preferred. The energy limit for LR-115 is 4 MeV and all the progeny isotopes of radon and thoron emit alphas with energies greater than 5 MeV. The films are less influenced by the moderate humidity, heat and light.

Calibration experiments were carried out at the Bhabha Atomic Research Center to estimate the calibration factors separately for radon and thoron, in a set up of stainless steel chamber of 0.5 cubic meter volume. The calibration chamber has facility for attributing aerosols from an aerosol generator. The chamber has also got provisions for an online Lucas cell system in conjunction with an Alpha Guard for continuous measurement of radon gas concentration. The online system was compared with Alpha Guard readings while radon concentration inside the chamber was varied for different experimental conditions. Radon or thoron, as the case may be, was introduced into the chamber from separate sources of Pylon, Canada^(16, 17). Aerosol generator used is a Sinclair LaMer type condensation aerosol generator, which gives a laminar flow of monodispersed aerosol of di-2-ethylhexyl sebate condensed on NaCl nuclei. The temperature settings of the boiler and reheaters are adjusted to obtain monodispersed aerosols of size 0.125 μm diameter. Aerosol concentrations of the order of 10^4 to 10^5 particles cm^{-3} of air were generated to stimulate the Indoor environmental conditions. The activity median aerodynamic diameter of the particles is referred to as particle diameter and is estimated to be 0.2 μm for Indoor aerosols. To maintain the equilibrium factors at the desired levels, the aerosols were continuously introduced. Decay pattern of the aerosols inside the chamber was studied and accordingly aerosols were put into the chamber to maintain the number concentrations. Alpha Guard kept inside the chamber records the hourly average radon concentrations. The online Lucas cell system was

coupled to an alpha counting setup and counts were taken synchronizing with the timing of the Alpha Guard. The comparison of radon measured by the two systems for a wide range of concentrations, showed very good correlation of 97% with a slope of unity (18, 19).

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 dimension with length 45 mm and radius 31 mm. The films used in the dosimeter are of approximately 12 μm thickness. The SSNTD-1 placed in compartment-A measures only radon, which diffuses into the compartment from the ambient air through a semi-permeable membrane (20). These membranes have permeability constants in the range of 10⁻¹² – 10⁻¹¹ m² 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 (21-23). On the opposite side, the glass fiber filter paper in the compartment-B allows both radon and thoron gas to diffuse in and hence the tracks on SSNTD-2 are measure of concentration of both gases. The SSNTD-3, compartment - C, exposed in the bare mode registers alpha tracks attributable to the air borne concentrations of both the gases and their progeny.

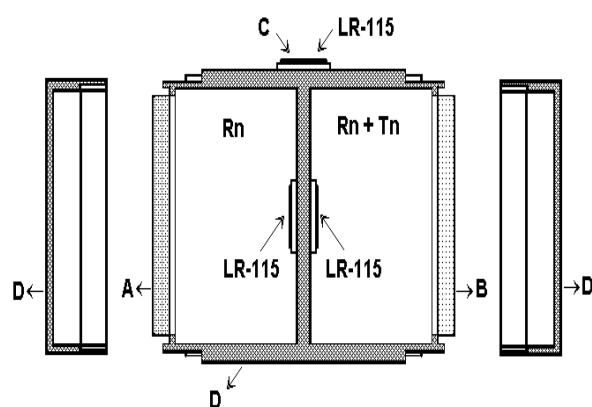


Figure 1. The double chamber dosimeter cup where A-filter paper and membrane, B- filter paper, C-bare detector and D- perforated covers.

These dosimeters are suspended at the centre of each room with different floor materials, wall materials and in different rooms such as bath room, kitchen, bed room, living room. This procedure is adopted for several houses in same location and also covering different areas. At the end of the stipulated period of exposure, usually 90 days, the dosimeters are retrieved and all the three SSNTD's are etched with 10% NaOH solution for one hour where the temperature of the bath is maintained at 60 °C (24).

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 and their progeny working level concentrations are calculated by the following relations (24):

$$C_R \text{ (Bq m}^{-3}\text{)} = T_m / (d \sigma_m) \quad (1)$$

$$C_T \text{ (Bqm}^{-3}\text{)} = (T_f - d C_R \sigma_{rf}) / (d \sigma_{rf}) \quad (2)$$

$$R_n \text{ (mWL)} = (C_R F_R) / 3.7 \quad (3)$$

$$R_T \text{ (mWL)} = (C_T F_T) / 0.275 \quad (4)$$

$$F_R = 0.104 f_{RA} + 0.518 f_{RB} + 0.37 f_{RC} \quad (5)$$

$$F_T = 0.91 f_{TB} + 0.09 f_{TC} \quad (6)$$

Where:

T_m is the track density of the film in membrane compartment

d is the period of exposure (in days)

σ_m refers to the sensitivity factor of membrane compartment

T_f is the track density of the film in filter compartment

σ_{rf} is the sensitivity of ²²²Rn in filter compartment

C_R and C_T is the concentration of ²²²Rn and ²²⁰Rn respectively

R_n and R_T refers to the progeny concentrations of ²²²Rn and ²²⁰Rn respectively

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

F_R and F_T represents the equilibrium factors for ²²²Rn and ²²⁰Rn progeny corresponding to the extracted ventilation rate .

The inhalation dose ($\text{mSv}\cdot\text{y}^{-1}$) is estimated by making use of equilibrium factor that was determined by the working level concentrations as ⁽¹⁴⁾:

$$D = 0.007 \times [(0.17 + 9F_R) C_R + (0.11 + 32F_T) C_T] \quad (7)$$

Estimation of Activity of ²²⁶Ra

The activity of ²²⁶Ra in the soil and building materials is estimated by using gamma ray spectrometer. The soil samples (about 10) were collected at different locations near the houses, where the houses are of different types of constructions. Like this almost all areas of Mysore city is covered. The places that are free from surface run-off during heavy rain were carefully selected. An area of about 0.5 m² was chosen, the vegetation and roots were removed, if any. Then about 2 kg of soil samples were collected from these places nearly at a depth of ~ 0.2 m from the earth's surface. 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 ²²⁶Ra, ²²²Rn and its progenies.

The spectrometer consisted of n-type Hyper Pure Germanium detector of good efficiency having a resolution of 1.75 keV at 1.33 MeV and 641 eV at 5.9 keV and coupled to a Multichannel Analyzer of 4096 channels. The detector is housed in a lead shield to screen the detector from the background radiation.

The gamma spectrometric procedure is followed to estimate the activity of ²²⁶Ra. The gamma peak of energy 609.51 keV that is emitted by ²¹⁴Bi, a decay product of ²²⁶Ra,

with intensity of 46.1% is used as proxy for the quantitative determination of ²²⁶Ra by gamma spectrometry. The activity concentration (Bq kg^{-1}) of radionuclide is estimated using the relation ^(25, 26):

$$\text{Activity concentration} = (S \pm s) 10^7 / (E W A) \quad (8)$$

Where:

S is the net counts per second under the photo peak

s is the standard deviation of S

E is the counting efficiency (in percent)

W is the mass of the sample (kg)

A is the gamma abundance (in percent) of the radionuclide

Indoor ²²²Rn Exhalation rate and concentration

The collection chamber method has been used for the measurement of ²²²Rn from indoor floor of different surfaces and in conjunction with the LLRDS. The collection chamber is made of stainless steel and appears as dome shaped vessel of 440 mm diameter, height of 100 mm and an effective volume of 15 liters. The time of collection of radon has been optimized for one hour. The radon concentration is estimated using LLRDS chamber and hence exhalation rate is measured by the concentration of radon. The exhalation rate from the soil is calculated by knowing the concentration of radon and using the expression ⁽²⁷⁾:

$$J = \frac{(V + v) C_R \lambda}{A [1 - \exp(-\lambda t)]} \quad (9)$$

Where:

J is the exhalation rate ($\text{Bq m}^{-2} \text{s}^{-1}$)

A is the exhalation area (m^2)

V is the volume of the accumulation chamber (m^3)

v is the volume of the LLRDS chamber (m^3)

λ is the decay constant of radon (s^{-1})

C_R is the concentration of radon in the collection chamber (Bq m^{-3})

t is the duration of accumulation of radon gas in the chamber.

RESULT AND DISCUSSION

Continuous measurements have been made using SSNTDs for both Indoor radon and thoron, simultaneously. Radon, thoron and their progeny concentrations were measured in different types of buildings 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 figures. 2-4. The variations may be due to different types of materials used for construction of the houses and also due to the ventilation rate.

Sathish *et al.* (28) have given the preliminary results of the diurnal and seasonal variations of the concentration of radon, thoron and their progenies at various locations in Mysore city. The authors have reported the arithmetic mean values of concentrations (Bq m^{-3}) of radon and thoron for Indoor atmosphere which ranged from 9.20 to 58.02 with a median of 34 and 7.21 to 59.27 with a median of 33, respectively. The arithmetic mean of progeny concentrations (mWL) varied from 0.03 to 2.52 with a median of 1.29 and 0.07 to 47.04 with a median of 20.70, respectively. The reported dose to the selected population of Mysore city was 1.58 mSv y^{-1} .

Different floorings

Figure 2 is the representation of variation of radon and thoron concentrations in different types of buildings at various locations in Mysore city. The houses for measurements were carefully chosen in such a way that the walls and flooring of the houses are of different materials. From the figure, higher concentration of radon is observed in granite floorings than other materials. The data shows that in the houses with brick and cement-flooring, concentrations of radon is less but in the house with granite flooring it is slightly higher because granite may contain higher concentrations of ^{226}Ra . Activity concentration of radium in different building materials is shown in table 1. Granite is

rich of radium and it may be the reason for higher concentration of radon in granite flooring houses. The materials used for construction of buildings are sufficiently porous and allow radon to enter into the indoor atmosphere (29). The earlier studies have shown that there is a positive correlation between the exhalation rate and concentration in building materials (30). Granite samples show higher radon exhalation rate than marbles. There is a positive correlation between radium content of granite with radon exhalation and its concentration. This trend is observed by several researchers (31-33).

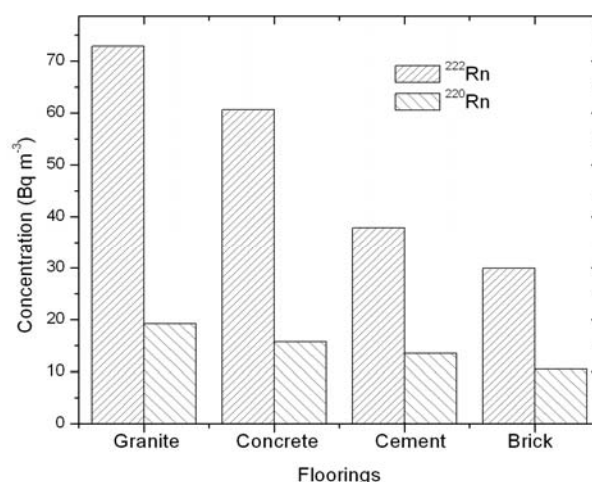


Figure 2. Variations of radon and thoron in different floorings.

Table 1. Activity concentration of ^{226}Ra in building materials.

Building materials	Activity concentration of ^{226}Ra (Bq kg^{-1})
Soil	4.0 ± 0.20
Mud	5.4 ± 0.27
Brick	12.5 ± 0.63
Cement	25.6 ± 1.30
Concrete	26.8 ± 1.34
Granite	54.5 ± 2.73

Some of the huts observed have mud walls, bare flooring and the roof covered by the dry coconut leaves with poor ventilation. Table 2 summarizes the exhalation of radon in different floorings. The exhalation from brick wall with cement plastering is high. It is observed that the Mosaic flooring houses

show slightly less radon exhalation rate than cement flooring houses. Radon concentration in huts is lower than in concrete and mosaic flooring houses even though radon exhalation from bare flooring is higher than other types of floorings. This may be attributed to the possibility of out flow of air from huts through the coconut dry leaves. Further, the fumes emitted during cooking process either from the kerosene stove or fire wood will increase the air pressure inside dwellings. Radon exhalation in mud walls is less compared to the cemented walls. High exhalation of radon from concrete walls may be responsible for enhancement of concentration in cement and mosaic flooring houses, due to large extent of usage of these materials in construction. This may be due to the high radon concentration in the concrete floorings and also because of the existence of porous between the slabs that were sandwiched by cement for the floorings. Due to this radon could easily enter into the houses ⁽³⁴⁾. The cracks in the floor as a consequence of poor quality of materials used for construction favours the enhancement of radon concentration ⁽³⁵⁾.

Sreenath Reddy *et al.* ⁽³⁶⁾ and Vinaya Kumar *et al.* ⁽³⁷⁾ have observed that the dwellings with mud floors have relatively higher dose and reports that the sub surface soil may be predominating source of indoor radon, thoron for the environment of

Hyderabad, India. This may be attributed to the rich content of radium and thorium in the local soil used for construction of mud walls. The radon levels in mud houses may be high due to emanation from ground surfaces and poor ventilation of the dwellings ⁽³⁸⁾. Radon concentrations in dwellings changes from place to place due to difference in geology and climate, in construction materials techniques and domestic customs. Although exposure for radon in tropical climates is unlikely to be of serious concern, keeping in view the radiological significance of radon, mapping of radon level in different parts of the country is helpful in defining the radon prone areas, if any, and evaluating the natural radiation exposures ⁽³⁹⁾.

Different walls

The average concentrations of radon and thoron levels for different types of wall are shown in figure 3. The concentrations 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 houses ⁽⁴⁰⁾. The reported average values of porosity of the soil and building materials are 0.25 and 0.15, respectively ⁽⁴¹⁾. The radon exhalation rate is higher in soil than in concrete, whereas building may contain several tons of concrete. The contribution of this to indoor radon is relatively low because of low

Table 2. ²²²Rn exhalation rate in different floorings.

Types of Surface	²²² Rn Exhalation rate (m Bq m ⁻² S ⁻¹)
<i>Ground Floor</i>	
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
<i>Wall Thickness(m)</i>	
0.36	3.85±0.62
0.09	0.75±0.14
Brick Wall	5.49±1.00

escape rate of radium. Radon exhalation rate also depends on the thickness of wall as evident in table 2. Samuelson⁽⁴²⁾ measured the radon exhalation rate in the walls and reported $20 \text{ Bq m}^{-2}\text{s}^{-1}$ for the dwellings of Scandinavian. The results are in comparison with the measured activity of radium for the building materials as shown in table 1. UNSCEAR⁽¹⁴⁾ has reported exhalation rates from walls and floor of half slab thickness $0.1\text{m} - 0.05\text{m}$ as $1.6 \text{ mBqm}^{-2}\text{s}^{-1}$ and $0.8 \text{ mBqm}^{-2}\text{s}^{-1}$, respectively. Ramachandran *et al.*⁽⁴³⁾ have quantified these results and reported the exhalation rate of ^{222}Rn from building materials. The reported values for concrete and brick vary from $0.43\text{-}1.3 \times 10^{-5}$ and $0.1 \times 10^{-5} \text{ Bqkg}^{-1}\text{s}^{-1}$ for USA, 0.47×10^{-5} and $0.017 \times 10^{-5} \text{ Bqkg}^{-1}\text{s}^{-1}$ for Denmark and 0.32 and $0.16 \times 10^{-5} \text{ Bqkg}^{-1}\text{s}^{-1}$ for USSR. The reported values for Hungary are 0.78×10^{-5} and $0.39 \times 10^{-5} \text{ Bqkg}^{-1}\text{s}^{-1}$, respectively.

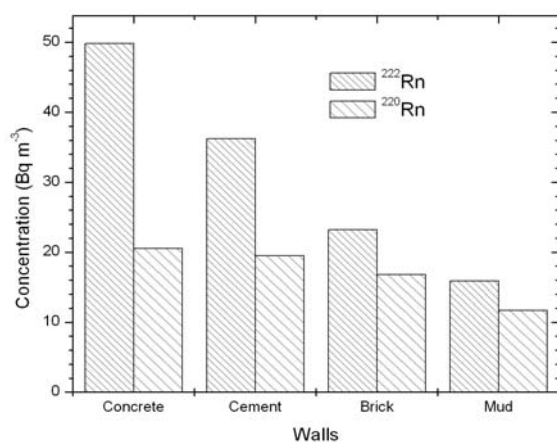


Figure 3. Variations of radon and thoron in different types of walls.

Different rooms

The figure 4 shows the annual average concentrations of radon and thoron 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 ^{222}Rn 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 ^{222}Rn due to ^{222}Rn dissolved in water⁽⁴⁴⁾. ^{222}Rn is shown to be released in spray from faucets or shower fixture^(45, 46). Air in living rooms on the other hand is most readily diluted due to outdoor air blow. This can be correlated with concentrations of ^{222}Rn in water⁽²⁸⁾ and the activity of ^{226}Ra . However, Yadagiri Reddy *et al.*⁽⁴⁷⁾ have reported that the radon concentration varied from 17 to 311 Bq m^{-3} and progeny concentration varied between 0.1 to 20mWL with an average of $62 \pm 45 \text{ Bq m}^{-3}$ and $4.7 \pm 4.1 \text{ mWL}$, respectively. They have also reported the activity of radium from different zones and showed the 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 and also depends on location of the houses for the environment of Hyderabad.

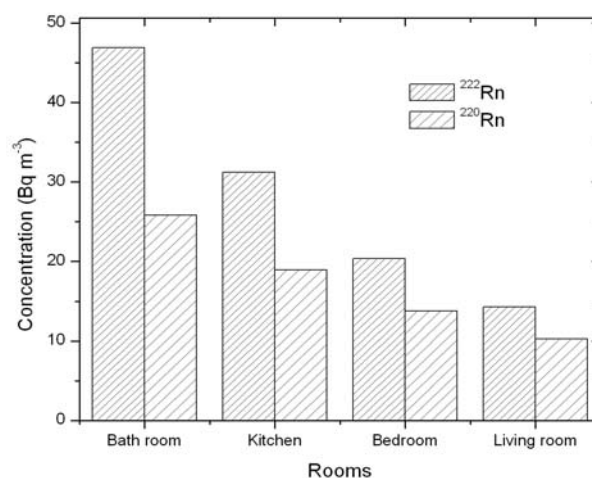


Figure 4. Variations of radon and thoron in different rooms.

CONCLUSION

One can conclude that the concentration level of radon and thoron in dwellings depend on various factors such as soil beneath, local geology, the house construction materials, microclimatic 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 radium rich materials to avoid the health hazards due to radon. Indoor radon and thoron concentration depends also on radon exhalation rate of the flooring and ventilation condition. Higher concentrations of both radon and thoron have been observed where the exhalation rate is more. The activity of radium in the soils of Mysore region is lower than the global average value. Indoor radon and thoron concentrations at Mysore are comparable with the global average value; the global average values being 40 and 10 Bq m⁻³, respectively.

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