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Activity of radium, thorium and potassium in soil and building materials

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ABSTRACT

The activities of ²²⁶Ra, ²³²Th and ⁴⁰K in soil samples and building materials have been carried out using Hyper Pure Germanium gamma ray detector. It is observed that the ²²⁶Ra and its progeny concentrations are higher in the houses having granite flooring. Activity concentration of ²²⁶Ra is measured by collection chamber method and the exhalation rate is done using solid state nuclear track detectors. A good correlation is observed for ²²⁶Ra and ²³²Th activity in soil with indoor ²²²Rn, ²²⁰Rn and its progeny in dwellings.

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KEYWORDS

Radium;
Thorium;
Radon;
Thoron;
Soil;
Building materials;
Indoor environment.

1. INTRODUCTION

Radon and its daughters are identified as a cause of lung cancer among the miners. It has not been clear whether radon poses a similar risk of causing lung cancer in humans exposed at generally lower levels found in homes, number of indoor radon survey have been carried out in recent years around the world reveals this truth. The assessment of radiological risk related to inhalation of radon and its progeny is based mainly on integrated measurements of radon. Generally the thoron concentration in dwellings is considered negligible because of the short half life of the thoron (55.6 s) and the world wide average estimated is 3 Bq m⁻³[1]. While calculating the risk, the contribution of thoron and its progeny can not be ignored. As such the radon measurements should be accompanied by the thoron measure-

ment for the assessment of the correct level of dose due to radionuclides present in the environment. Several researchers^[2-6] have studied the effect of radon/thoron on human beings. Radon exposures are largely determined by the geological underlying the buildings, its construction and ventilation conditions^[7]. The concentration of radon and its decay products show large temporal and local fluctuations in the indoor and outdoor atmosphere due to variation of temperature, pressure, building materials, ventilation condition, wind speed^[8-10] etc.

Measurement of indoor radon are important because of the radiation dose to human population due to inhalation of radon and its daughters contributes more than 50% of the total dose from natural sources^[11]. The three isotopes (²²²Rn, ²²⁰Rn, ²¹⁹Rn) are gaseous and they may be released from the ground, rocks and also

from building materials and accumulate with their short lived daughters in closed spaces, and particularly in dwellings. ^{220}Rn and ^{219}Rn are not as important as ^{222}Rn , mainly because of their short half-life. The later may reach higher levels of concentration in the air that are significant in terms of radiological protection. The dose derived from the presence of radon in the air is linked to the inhalation of its short-lived daughters, which are deposited in the respiratory organs, if deeply inhaled; emit alpha-particles in direct contact with the bronchial and pulmonary epithelium. For these reasons, the doses deriving from the exposures to radon in closed spaces have been placed in direct relation to the risk of lung cancer.

Measurements were made for the environment of Mysore city (12N, 76E) where the sand appears as red sandy loam. The soil being sufficiently porous and permits free internal and downward movement of water and also diffusion of radon into the atmosphere depending on water content. In the present investigation an attempt has been made to find the activities of ^{226}Ra , ^{232}Th and ^{40}K in soil samples and building materials, which are used for the construction of houses. Also made an attempt to study the correlation of ^{226}Ra and ^{232}Th activities in soil with indoor ^{222}Rn , ^{220}Rn and its progeny in dwellings.

2. EXPERIMENTAL

2.1. Solid state nuclear track detectors (SSNTD's)

The concentrations of radon, thoron and their progeny are measured in dwellings using Solid State Nuclear Track Detectors (SSNTD), which are thin sheets of dielectric materials such as cellulose nitrate (CN) and polycarbonate. SSNTDs are sensitive to alpha but not to beta and gamma radiation. An alpha particle passing through such an insulating media will leave narrow (3-10 nm) trail of damage. This damage produces broken molecular chains, free radical etc. These damaged regions can dissolve at a much higher rate than the undamaged material in certain chemical agents called etchants. The dissolved portion appears as a track in the film. The number of such tracks gives the number of alpha particles. The dosimeters are constructed using SSNTDs. The dosimeter used is a cylindrical plastic

cup divided into two components having a provision for holding the SSNTD films in specific concentration. The SSNTD is a cup with a suitable membrane like thin latex rubber sheet that determines the ^{222}Rn concentration alone since ^{220}Rn gas is trapped to less than 1% and SSNTD inside the cup with a filter paper determines the both ^{222}Rn and ^{220}Rn concentrations in air. The system used for these measurements is called double chamber dosimeter. The bare mode exposure film can be fixed conveniently on the surface of the chamber. For indoor measurements normally LR-115 TYPE II (Kodak Pathe, France) plastic track detectors are preferred^[12-15].

The schematic representation of the double chamber dosimeter cup used in the study for monitoring radon, thoron and their progeny is shown in figure 1. The chamber has cylindrical in shape with 45 mm and 62 mm of length and diameter, respectively. 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 it from the ambient air through a semi-permeable membrane such as latex, cellulose, nitrate etc. These membranes have good permeability of the order of 10^{-12} - 10^{-11} $\text{m}^2 \text{s}^{-1}$ and allow more than 95% of radon gas to diffuse, and reduce thoron concentration to the extent of 1% or even less^[16]. 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 the gases. The SSNTD-3 exposed in the bare mode that is placed on the outer surface of the dosimeter registers alpha tracks attributable to the airborne concentrations of both the gases and their progeny^[17]. These dosimeters are suspended from

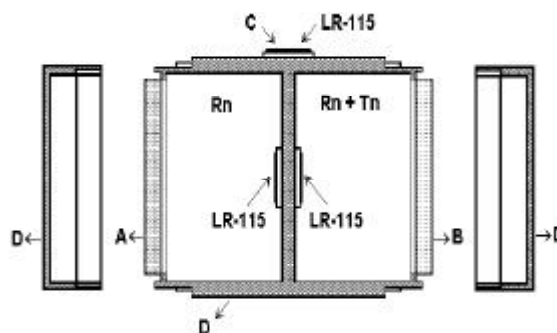


Figure 1: Embedded view of twin cup dosimeter

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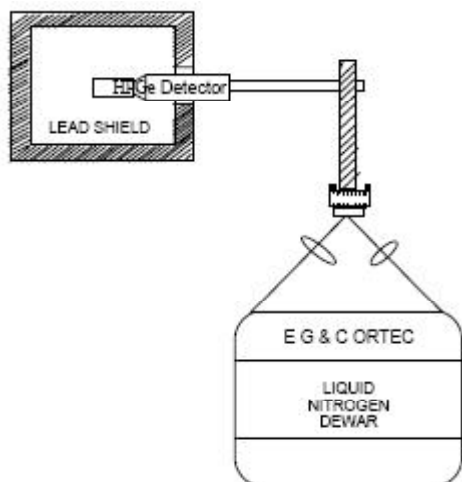


Figure 2: Block diagram of HPGe gamma spectrometer

the midpoint of the house at a height of approximately 2 m from the floor of the dwelling. 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^[18]. 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 estimated by the following relations^[17].

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

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

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, and, 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.

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

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

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 correspond-

ing to the extracted ventilation rate^[17]. Equilibrium factor is determined using the working level concentrations, and the inhalation dose^[19] (m Sv y^{-1}) is estimated by using:

$$D = 7 \times [(0.17 + 9F_R) C_R + (0.11 + 32F_T) C_T] \times 10^{-3}$$

2.2. Estimation of activity of ^{226}Ra , ^{232}Th and ^{40}K

The activities of ^{226}Ra , ^{232}Th and ^{40}K in the soil and building materials are measured using gamma ray spectrometer. The schematic representation of Hyper Pure Germanium gamma ray detector (HPGe) set up is shown in figure 2. 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 run-off during heavy rain were carefully selected. An area of about 0.5 m² 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 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^[20,21]. 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 having a resolution of 1.75 keV at 1.33 MeV and 641 eV at 5.9 keV and coupled to a 4096 channel Multi Channel Analyzer. The detector is housed in a lead shield to cease the background radiation entering from outside. This type soil sampling was aimed at evaluating the distribution of radionuclides in the surface soil to correlate the same with radiation exposure level and the availability of radionuclides that can be picked up by vegetation growing on the soil.

The gamma spectrometric procedure is followed to estimate the activity of radium. The gamma peak of energy 609.51 keV (which is emitted by ^{214}Bi , a decay product of ^{226}Ra) with intensity of 46.1% is used as proxy for the quantitative determination of ^{226}Ra by gamma spectrometry^[22]. The activity (Bq kg^{-1}) of ra-

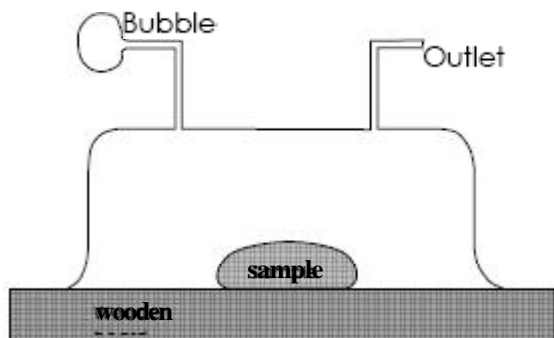


Figure 3: Schematic of the emanation of radon from samples of building materials

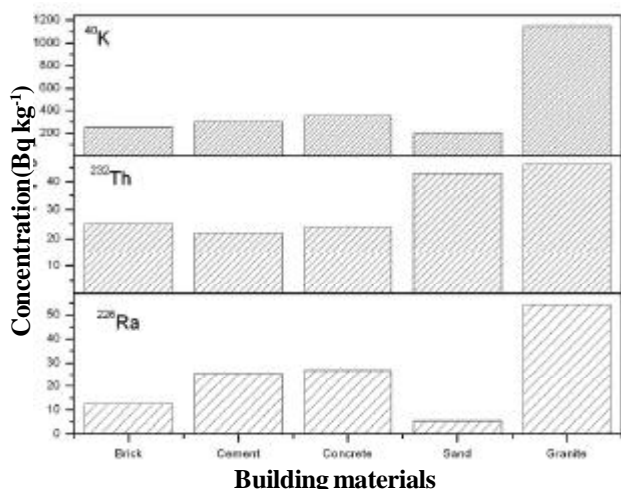


Figure 4: Variation of ²²⁶Ra, ²³²Th, ⁴⁰K in building materials

dionuclide is calculated using the relation:

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

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 and W is the mass of the sample (g).

2.3 Indoor ²²²Rn exhalation rate and concentration

The collection chamber method has been used for the measurement of ²²²Rn from indoor floor of different surfaces. Measurements were made using Low Level Radon Detection System (LLRDS). The procedure for the measurement of ²²²Rn exhalation rate involves of the following steps. First, collecting the ²²²Rn exhaled from a known area of the indoor floor for a known time in a collection chamber. Transfer of a specific fraction of the air from the collection chamber to LLRDS^[23]. The ²²²Rn concentration is estimated using LLRDS chamber and hence ²²²Rn exhalation rate is measured

by the concentration of radon.

The schematic representation of collection chamber setup, for the measurement of ²²²Rn exhalation rate in indoor floor is shown in figure 3. The accumulation chamber is a dome shaped stainless steel vessel with the rim diameter of 440 mm and a height of 100 mm having an effective volume of 15 liters. On the top of the chamber, two openings are provided one for connecting a hard rubber bulb which is used for mixing the air uniformly in to the collection chamber and the other for transferring the air from collection chamber to the LLRDS. Any leakage of air through the sides at the bottom of the chamber is avoided by proper sealing by sealant. ²²²Rn exhaled from the floor gets collected in the chamber. The time of collection of ²²²Rn for transferring it to LLRDS has been optimized for one hour. The exhalation rate from the soil is calculated by knowing the concentration of radon and using the following expression:

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

where J is the exhalation rate (Bq m⁻² s⁻¹), A is the exhalation area (m²), V refers to the volume of the accumulation chamber (m³), v is the volume of the LLRDS chamber (m³), λ stands for the decay constant of radon (s⁻¹), C_R is the concentration of ²²²Rn in the collection chamber (Bq m⁻³) and t is the duration of accumulation of radon gas in the chamber.

3. RESULTS AND DISCUSSION

The figure 4 shows the mean values of ²²⁶Ra, ²³²Th and ⁴⁰K in different building materials. The results reveals that the activity concentrations are relatively more in granite than the other materials used for the construction of the houses and may lead for the higher concentrations of radon/thoron in dwellings. The radium content is found maximum in granite and minimum in sand compared to cement, concrete and brick. The activity of thorium is more in granite and sand and in other materials such as cement, concrete and brick it appears to be more or less same. The results obtained are of the same order as observed by others^[24-26]. The variation of ⁴⁰K is also observed to see the activity contribution. The potassium activity is found more in granite compared to the other materials used for the construction. The samples were carefully selected for ⁴⁰K in such a

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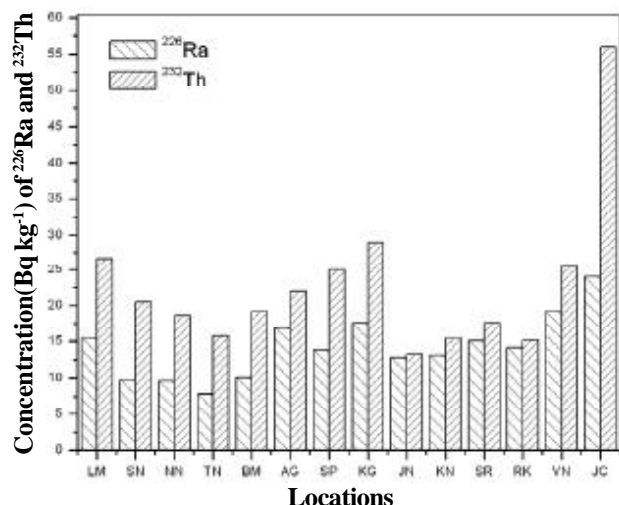


Figure 5: Concentrations of ^{226}Ra and ^{232}Th in different locations (LM-Lashkar Mohhala, SN- Siddique Nagar, NN-Naidu Nagar, TN-Tilak Nagar, BM-Bada Makhan, AG-Agrahara, SP-Saraswathipuram, KG-KG Koppal, JN-Jayanagar, KN-Kuvempu nagar, SR-Srirampura, RK-Ramakrishnanagara, VN-Vivekananda Nagar, JC-J C Nagar)

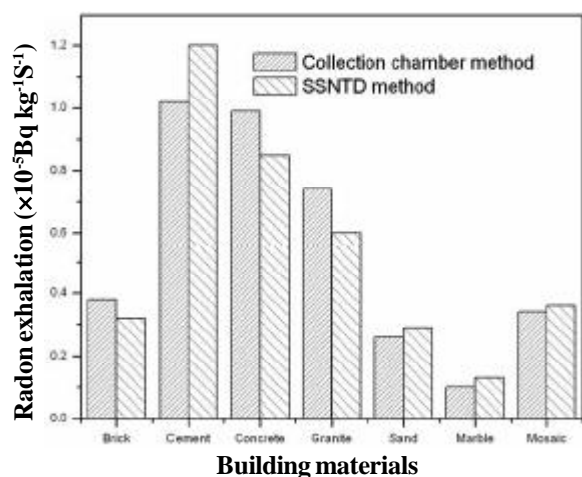


Figure 6: Comparison of radon exhalation from collection chamber and SSNTD methods

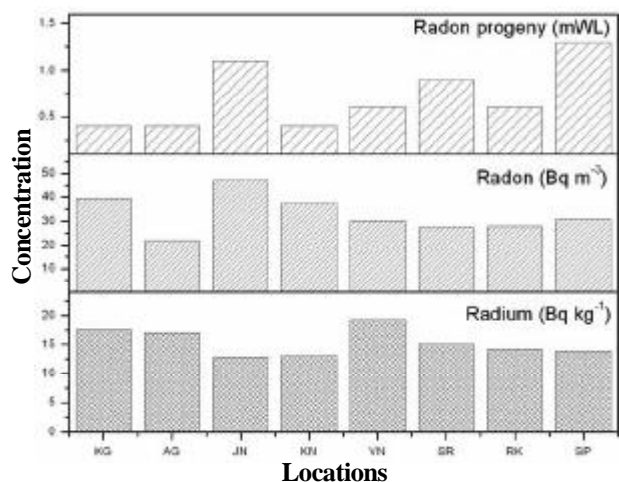
way that the locations are purely vegetation land, where the radium/thorium content is negligible. Extensive works on concrete building blocks were carried out by Ademola and Oguneletu^[27] for Ibadan in Nigeria. They have used the same methodology as described in this paper for the estimation of activity concentration of radium, thorium and potassium using gamma ray spectrometry. The radioactivity concentrations were found to vary from 6.2 to 57.5 Bq kg⁻¹, 12.4 to 64.9 Bq kg⁻¹ and 95.3 to 766.1 Bq kg⁻¹. El Bahi^[28] has reported the

average values of ^{238}U , ^{232}Th , and ^{40}K activity concentrations for different types of cement that are lower than the corresponding global values. The effective radium content and the exhalation rate are found to vary from 12.75 to 38.52 Bq kg⁻¹ and 61.19 to 181.39 Bq m⁻² d⁻¹, respectively. Rohit et al.^[29] have measured the activity concentrations of soil samples of thirty different locations of Malwa region of Punjab by using HPGe detector. The range of activity concentrations mentioned for ^{226}Ra , ^{232}Th and ^{40}K in the soil varies from 18.37 Bq kg⁻¹ (Sangrur) to 53.11 Bq kg⁻¹ (Sitoguno), 57.28 Bq kg⁻¹ (Dhanola) to 148.28 Bq kg⁻¹ (Sitoguno) and 211.13 Bq kg⁻¹ (Sunam) to 413.27 Bq kg⁻¹ (Virk Khera) with overall mean values of 35 Bq kg⁻¹, 80 Bq kg⁻¹ and 317 Bq kg⁻¹, respectively.

The figure 5 shows the activity of ^{226}Ra and ^{232}Th in different locations. The radium content in soil range from 7.85 to 19.2 Bq kg⁻¹ with a mean of 13.51±3.4 Bq kg⁻¹ and for thorium it varies from 13.25 to 29.0 Bq kg⁻¹ with a mean of 20.29±4.9 Bq kg⁻¹, excluding the area JC Nagar, in both the cases. The highest activity of ^{226}Ra and ^{232}Th in JC Nagar may be attributed to the granite rocks surrounding the area. Activity concentration of ^{232}Th is higher than ^{226}Ra at all location because the soil and rocks of Mysore region is surrounded by pegmatite and granite rocks^[21]. Relatively JC Nagar shows highest compared to the other locations. The ratio of ^{232}Th and ^{226}Ra can be used as an indicator of the relative abundance of ^{232}Th and ^{238}U . The ratio ranges from 1.04 to 2.33 with a geometric mean of 1.6. The ratio is always greater than unity and envisages the more abundance of thorium compared to uranium in these areas. The average values of ^{226}Ra and ^{232}Th reported for normal background areas of Indian soils are 15 and 18.36 Bq kg⁻¹, respectively^[30]. Kant et al.^[31] reports the concentration of Radium, Thorium and Potassium in the mixed soil sample from crop fields as 16.2±0.22, 68.1±1.44 and 875.0±9.68 Bq kg⁻¹, for barren soil sample 9.1±0.13, 59.4±1.45 and 668.4±8.01 Bq kg⁻¹ respectively. The radium equivalent activity in the soil sample from crop fields is 225.9 Bq kg⁻¹ and in barren soil sample it was 193.1 Bq kg⁻¹. The activity of ^{226}Ra is compared with the radon exhalation rate by different techniques such as collection chamber and solid state nuclear track detector method. This variation is depicted in bar diagram shown

TABLE 1: Annual average radon, thoron and their progeny concentrations and equilibrium factor

Locations	Concentration (Bq m ⁻³)		Progeny concentration (mWL)	
	²²² Rn	²²⁰ Rn	²²² Rn	²²⁰ Rn
	Lashkar Mohalla (LM)	12.9	18.8	0.610
Siddiquenagar (SN)	10.1	9.0	0.254	0.041
Naidunagar (NN)	17.4	28.3	0.246	0.131
Mandimohalla (MM)	12.6	20.7	0.400	0.094
Tilaknagar (TN)	9.3	21.2	0.013	0.001
Badamakkan (BM)	9.2	33.1	0.706	0.391
NR Mohalla (NM)	12.5	18.9	0.360	0.196
Agrahara (AG)	40.7	45.5	0.013	0.001
Saraswathi puram (SP)	44.4	38.6	0.014	0.001
KG Koppal (KG)	54.5	42.1	0.013	0.001
Jayanagar (JN)	30.2	20.3	0.017	0.001
Kuvempu nagar (KN)	26.7	22.1	0.062	0.005
Sriramapura (SR)	30.0	19.2	0.014	0.001
R K Nagar (RK)	34.0	19.3	0.026	0.002
Vivekanandanagar (VN)	37.0	27.6	0.016	0.001
JC Nagara (JN)	58.0	40.2	0.025	0.002

**Figure 7: Concentration of radium, radon and its progeny in different locations**

in figure 6. The exhalation rate is relatively high in cement and concrete compared to the other materials. This may be attributed to the large usage of cement in the construction of buildings and hence the buildings may contain several tons of concrete compared to the other materials. The radon exhalation rate ranges from 1 to 10.2 μ Bq kg⁻¹ s⁻¹ with a mean of 5.5 μ Bq kg⁻¹ s⁻¹ for collection chamber method. While in the solid state nuclear track detector method, the exhalation rate varies from 1.3 to 12.0 μ Bq kg⁻¹ s⁻¹ with a mean of 5.4 μ Bq kg⁻¹ s⁻¹. In both the methods the minimum concentration was observed for marble and maximum is found

in cement. The results show a positive correlation between the two methods with a correlation of 95%. The annual average of indoor radon, thoron and their progeny concentrations for sixteen locations are shown in TABLE 1. The radon and thoron varies from 9.2 to 58 Bq m⁻³ with a mean of 27.4 Bq m⁻³, and 9.0 to 45.5 Bq m⁻³ with a mean of 26.5 Bq m⁻³, whereas their progenies varies between 0.013 to 0.7 mWL with a mean of 0.17 mWL, and 0.001 to 0.4 mWL with a mean of 0.07 mWL. The average values of radon concentration in villages viz. Aghar, Chakmoh, Gallot, Khian and Rachaon of Himachal Pradesh, India is found to be 0.73, 0.66, 1.06, 0.88 and 0.71 kg Bq m⁻³ [32]. Sanada et al. [33] have found the mean annual mean indoor radon concentration of 15.5 Bq m⁻³ for Japan. Virk and Sharma [34] have reported the average radon concentration the environment of Himachal Pradesh. The observed radon concentration ranges from 19.7 to 146.3 Bq m⁻³ while for thoron it is 9.1 to 70.7 Bq m⁻³. Singh et al. [35] have reported the values radon concentrations of 156.11 and 635.42 Bq m⁻³ for the dwellings of Kasol and Balsari villages in India, respectively. The values obtained from our measurements are comparable to the observation made elsewhere. The concentrations and distribution of natural and anthropogenic radio nuclides in soils from around the Kestanol (Çanakkale), Turkey were investigated by Merdanoglu and Altinsoy [36] to assess the environmental radioactivity and radiological health hazards. They also found that the concentration of ²³²Th in soil was higher than that of the ²³⁸U and the activities of ²³²Th and ²³⁸U in this area are higher than the world average. The results show that the mean radium equivalent activity is 498 Bq kg⁻¹.

The vertical bar graph in figure 7 shows the variation of radium, radon and radon progeny concentrations at different locations. The activity of radium varies between 12.8 and 19.2 Bq kg⁻¹ with a mean of 15.4 \pm 2.3 Bq kg⁻¹. In these locations the indoor radon ranges from 21.8 to 49.2 Bq m⁻³ with a mean of 32.8 \pm 8.0 Bq m⁻³ and its progeny ranges from 0.4 to 1.3 mWL with a mean of 0.72 \pm 0.3 mWL. The maximum activity of radium is found in Vivekanandanagar, Agrahara and KG Koppal. The higher amount of ²²⁶Ra in soil depends to a large extent on the mineral composition of the host rocks. The rocks in this belt are of rich in granite, and usually granite has large amount of radium than

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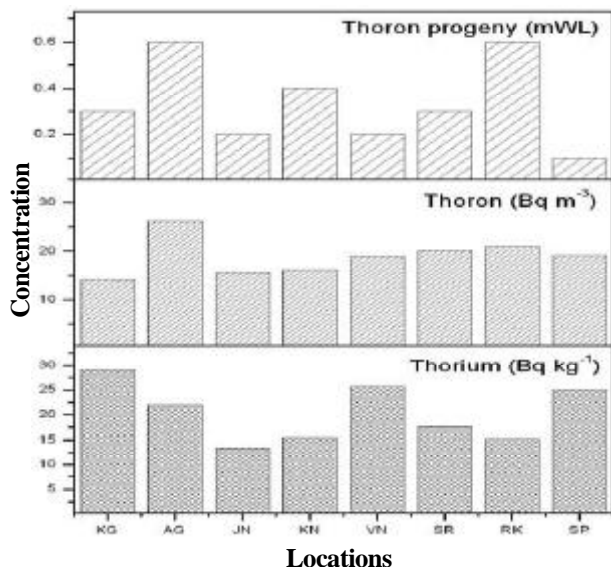


Figure 8: Concentration of thorium, thoron and its progeny in different locations

other rocks. The ^{226}Ra activity does not vary significantly in other parts of the city.

Figure 8 is the representation of the activity concentration of thorium. The concentration of thorium varies between 13.3 and 29.0 Bq kg^{-1} with a mean of $20.4 \pm 5.7 \text{ Bq kg}^{-1}$. The figure also shows the variation of indoor thoron and its progeny. In these locations the thoron ranges from 14.0 to 26.1 Bq m^{-3} with a mean of $18.9 \pm 3.7 \text{ Bq m}^{-3}$ and its progeny ranges from 0.1 to 0.6 mWL with a mean of $0.34 \pm 0.2 \text{ mWL}$. The maximum activity of thorium is observed at KG Koppal, Vivekanandanagar and Saraswathipuram. Activity concentration of ^{232}Th is higher compared to ^{226}Ra at all locations.

4. CONCLUSIONS

The radium is found to be maximum in granite and minimum in sand compared to cement, concrete and brick. The activity of thorium is more in granite and sand and in other materials such as cement, concrete and brick it appears to be more or less same. The activity of ^{226}Ra and ^{232}Th in JC Nagar is relatively high compared to other place and may be due to the presence of granite rocks surrounding the area. Activity concentration of ^{232}Th is higher than ^{226}Ra at all location because the soil and rocks of Mysore region is

surrounded by pegmatite and granite rocks. The higher amount of ^{226}Ra in soil depends to a large extent on the mineral composition of the host rocks. The rocks in this belt are of rich in granite, and usually granite has large amount of radium than other rocks. The ^{226}Ra activity does not vary significantly in other parts of the city. Activity concentration of ^{232}Th is higher compared to ^{226}Ra at all locations. The activity of ^{226}Ra and ^{232}Th are below the global average value. A good correlation between ^{226}Ra and ^{222}Rn and ^{232}Th and ^{220}Rn is also observed.

5. ACKNOWLEDGMENTS

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