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The natural radio- activity of some building materials used in sathanoor, Thiruvannamalai Dist, Tamilnadu, India.

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Abstract : Using γ spectrometry, the concentration of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K has been measured in Soil, Sand, Cement, Clay and Bricks, which are used as building materials in Sathanoor, Tiruvannamalai dist, Tamilnadu, India. The radium equivalent activity (Ra_{eq}), the criterion formula (CF), indoor gamma absorbed dose rate (D_R), annual effective dose (HR) external radiation hazard index (H_{ex}), internal radiation hazard index (H_{in}), associated with the natural radionuclides are calculated to assess the radiation hazard of the natural radioactivity in the building materials. From the analysis, it is found that these materials used for the construction of dwellings are safe for the inhabitants.

Keywords : Natural radioactivity, Building Materials, Gamma Ray spectrometry.

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1. Introduction : The values of specific radioactivities of soil and building materials are required in setting the standard and guidelines for their safe usage and in assessing the radiation hazards associated with them. All building materials such as concrete, cement, brick, sand, aggregate, marble, granite, limestone, gypsum etc contain mainly natural radionuclides mainly, including uranium (^{238}U) and thorium (^{232}Th) and their decay products and the radioactive isotope of potassium (^{40}K). In the ^{238}U series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important and, therefore, reference is often made to ^{226}Ra instead of ^{238}U. The naturally occurring radionuclides in the building materials contribute to radiation exposure, which can be divided into external and internal exposure. External exposure is caused by direct gamma radiation while internal exposure is caused by the inhalation of the radioactive inert gas radon (^{222}Rn, a daughter product of ^{226}Ra) and its short lived secondary decay products. Knowing the level of the natural radioactivity in building materials is important to assess the associated radiological hazards to human health and to develop standards and guidelines for the use and management of these materials.

The natural radioactivity of building materials in many countries has been reported (Ahmad and Matiullah Hussain, 1988; Amrani and Tahtat, 2001; Beretka and Mathew, 1985; Chong and Ahmad, 1982; Mollah et al., 1986; Viresh Kumar et al., 1999; Zaidi et al., 1999; El-Arabi, 2005; El-Tahaway and Higgy, 1995; Khalid Khan and HasanKhan, 2001; McAulay and Moran, 1988; Safdar Ali et al., 1996; Turhan, 2008; Stoulos et al., 2003; Tufail et al., 1992). Natural radioactivity in some Indian building materials has also been reported by other authors (Viresh Kumar et al., 1999; Ravisankar et al., 2012; Ajay Kumar et al., 2003; Nageswara Rao et al., 1996). However, detailed information for each state is scant. Data regarding the concentration of ^{226}Ra, ^{232}Th and ^{40}K in the building materials of Sathanoorr, Tiruvannamalai in the state of Tamilnadu in India is not available in the literature.

Sathanoor is a Village in Thandrampet Taluk in Tiruvannamalai District of Tamil Nadu State, India. It is located 17 KM towards west from District head quarters Thiruvannamalai and 201 KM from State capital Chennai. At Sathanoor on river Pennai beautiful dam has been erected and completed in 1964 to store water for the purpose of irrigating thousands of acres of vast stretch of dry belt of land. A mini hydel power plant is installed to add to the generation of power to the state of Tamilnadu. A beautiful Botanical garden has been raised on the sheltered lower side of the dam culturing varieties of ornamental plants and their flower adds charm to the spot. A zoological park of tourist interest is also set up there. It is very good picnic spot for weekenders as well as those who are visiting Tiruvannamalai. Transport services are available from Tiruvannamalai.

In the present work, the concentration of natural radionuclides was measured in 5 kinds of samples of building materials that were commonly used in Sathanoor, Tiruvannamalai, Tamilnadu, India, using gamma spectrometry. The potential radiological hazards associated with the studied materials were assessed by calculating the radium equivalent activity (Ra_{eq}), criteria formula (CF), indoor gamma absorbed dose rate (D_R), annual effective dose (H_R), external radiation hazard index (H_{ex}), internal radiation hazard index (Hin. The obtained results were compared to the recommended values to assess the radiation hazards to humans resulting from the building materials, and these results were also compared to the corresponding values of building materials from different countries.

2. Materials and methods

2.1. Sample collection and preparation

Commonly used structural building materials (clay, soil, brick, sand and cement) were collected randomly from sites where housing and other buildings were under construction and from building-material suppliers in Sathanoor, Tiruvannamalai for measurement of the specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K. These materials were used in bulk amounts. The materials were studied in their natural form. Each sample was properly cataloged, marked and coded according to its origin and the location of the sampling site.

After crushing, powdering, coning and quartering, representative samples with a maximum grain size of 1 mm were dried in an oven at approximately 110 C until the sample weight became constant. These samples were sealed in radon-impermeable plastic containers. The samples were then stored for more than 30 days to bring ^{222}Rn and its short-lived daughter products into equilibrium with ^{226}Ra (Ravisankar et al., 2012).

2.2. Gamma-ray spectroscopic technique

All selected samples were subjected to gamma spectral analysis using a 7.62 cm × 7.62 cm NaI (Tl) detector. The energy resolution of the NaI (Tl) detector measured in terms of the full width at half maximum (FWHM) is 50 keV at the energy of 662 keV gamma of 137Cs at 25 cm from the top of the detector. The detector is shielded with 15 cm thick lead on all sides including the top, to reduce the background contribution from the surroundings. The inner sides of the lead shielding are lined with 2 mm thick cadmium and 1 mm thick copper to attenuate lead X-rays and cadmium X-rays, respectively. The certified IAEA reference materials RGU, RG Th and RGK were used for the energy and efficiency calibration of the system in the energy range from

186.21 to 2614.53 keV. The activity contents of the IAEA reference materials, which are housed in 250 ml bottles, are known with73% accuracy. The efficiency percentages for 40K (1.461 MeV), 214Bi (1.764 MeV) and 208Tl (2.615 MeV) were found to be 0.154, 0.357 and 0.301 cps Bq^{-1}, respectively. The samples were sealed in radon-impermeable plastic containers.

The samples were then stored for more than 30 days to bring ^{222}Rn and its short-lived daughter products into equilibrium with ^{226}Ra. The samples were then counted in the same source-to-detector geometry used for the establishment of the efficiency calibration. The spectra were acquired for 20,000 s and the photo peaks were evaluated by the MCA software. The gamma-ray photo peaks corresponding to 1.461 MeV (40K), 1.764 MeV (214Bi) and 2.615 MeV (208Tl) were considered to determine the activities of 40K, 226Ra and 232Th in the samples. The detection limits of the NaI (Tl) detector system at the 95% confidence level for ^{40}K, ^{226}Ra and ^{232}Th are 8.50, 2.21 and 2.11 Bq kg^{-1}, respectively, for a counting time of 20,000 s. The results for the activity concentrations of the samples are reported with 2σ errors.

3. Results and discussion

3.1. Specific radioactivity

The activity concentrations of the detected radionuclides ^{226}Ra ^{232}Th and ^{40}K in these building materials are presented in Table 1. As shown in Table 1, the highest values observed for the specific activities of ^{226}Ra, ^{232}Th and ^{40}K are 31.26 Bq kg^{-1} (Cement), 198.58 Bq kg^{-1} (Sand) and 429.58 Bq kg^{-1} (Clay), respectively, while the lowest observed values of the specific activities of the same radionuclides are BDL, 20.9 Bq kg^{-1} (Soil) and 233.92 Bq kg^{-1} (Cement) respectively. As shown in Table 1, the activity of ^{226}Ra varies from BDL to 31.26 Bq kg^{-1} (Cement) and the arithmetic mean is 17.50 Bq kg^{-1}. The activity concentration of ^{232}Th varies from 20.9 Bq kg^{-1}

(Soil) to 198.58 Bq kg^{-1} (Sand), and the arithmetic mean is 64.03 Bq kg^{-1}. The activity concentration of ^{40}K varies from 233.92 Bq kg^{-1} (Cement) to 429.58 Bq kg^{-1} (Clay) and the arithmetic mean is 338.30 Bq kg^{-1}.

Table 1
Activity concentration and radiological parameters of building materials used in Sathanur, Tiruvannamalai dist, Tamilnadu.

Materials	Activity Concentration (Bq/kg)			Ra$_{(eq)}$ (Bq/kg)	Criteria Formula (mSvy^{-1})	Absorbed Dose Rate (D$_R$) (nGyn^{-1})	Annual Effective Doss H$_R$ (msvy^{-1})		Radiation Hazards	
	^{226}Ra	^{232}Th	^{40}K						H$_{ex}$	H$_{in}$
BRIC	4.16	27.45	294.60	66.09	0.08	57.59	0.28	0.07	0.156	0.312
CLAY	BDL	31.50	429.58	78.12	0.10	69.01	0.33	0.08	0.004	0.006
SAND	17.1	198.58	313.57	323.21	0.43	259.25	1.27	0.31	0.705	1.406
SOIL	BDL	20.90	419.86	62.21	0.08	56.57	0.27	0.06	0.003	0.006
CMNT	31.26	41.75	233.92	108.97	0.14	67.51	0.33	0.08	0.183	0.366
Average	17.50	64.03	338.30	127.72	0.17	101.99	0.49	0.12	0.210	0.419

The mean values are lower than the corresponding worldwide average values, which are 35, 30 and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively (UNSCEAR, 2000). If the radionuclide activities of the present study are compared to the world average values, the ^{226}Ra activity is lower by a factor of 0.29, while the activity of ^{232}Th is found to be higher by a factor of 1.63 and the ^{40}K activity is lower by a factor of 0.89. Figure-1 shows the activity concentrations of the natural radionuclides for the different building materials.

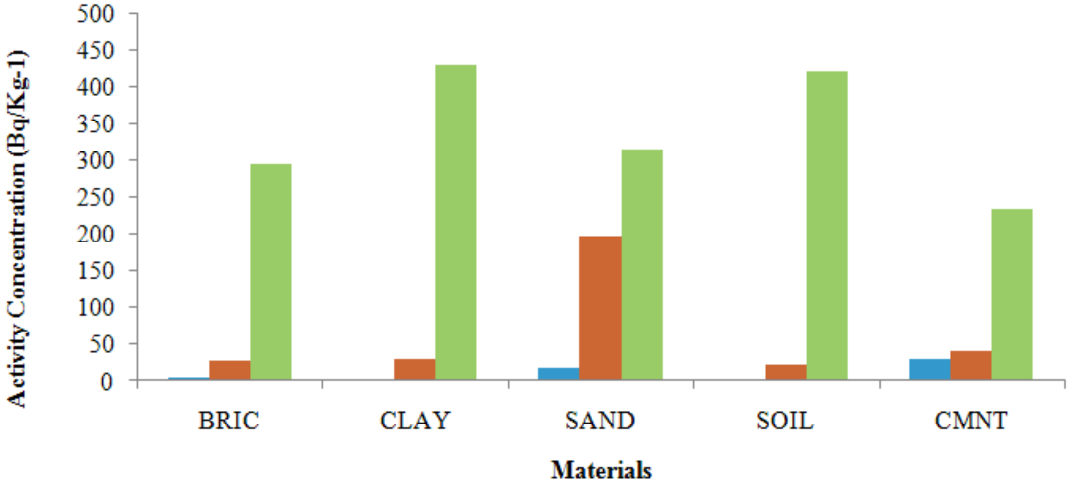

Figure 1 : Different types of Materials Vs Activity concentration.

3.2. Radium equivalent activity (Ra_{eq})

To represent the activity levels of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common radiological index has been introduced. This index is called radium equivalent (Ra_{eq}) activity and is mathematically defined (Beretka and Mathew et al, 1985) by

$$\mathbf{Ra_{eq}(Bq\ Kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K}} \text{ --------- } \mathbf{(1)}$$

where A_{Ra}, A_{Th}, A_K represents the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K. In the above relation, it has been assumed that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K produce equal γ- dose. The maximum dose Ra_{eq} in building materials must be less than 370 Bq kg^{-1} for safe use, i.e., to keep the external dose below 1.5 mGy yr^{-1} **(**Beretka and Mathew et al, 1985).

The calculated Raeq values range from 62.216 (Soil) to 323.214 (Sand) Bq kg^{-1} with an average of 127.725 Bq kg^{-1}. All values of Ra_{eq} in the studied samples are found to be lower than the criterion limit of 370 Bq kg^{-1} (NEA-OECD,1979). The results of this study show that the average value of Ra_{eq} obtained for the building materials is

127.725 Bq kg^{-1}, which is below the recommended maximum value (370 Bq kg^{-1}) and thus does not pose any radiological hazard when used for the construction of buildings. Figure 2 shows the building materials sample ID variation with radium equivalent activity.

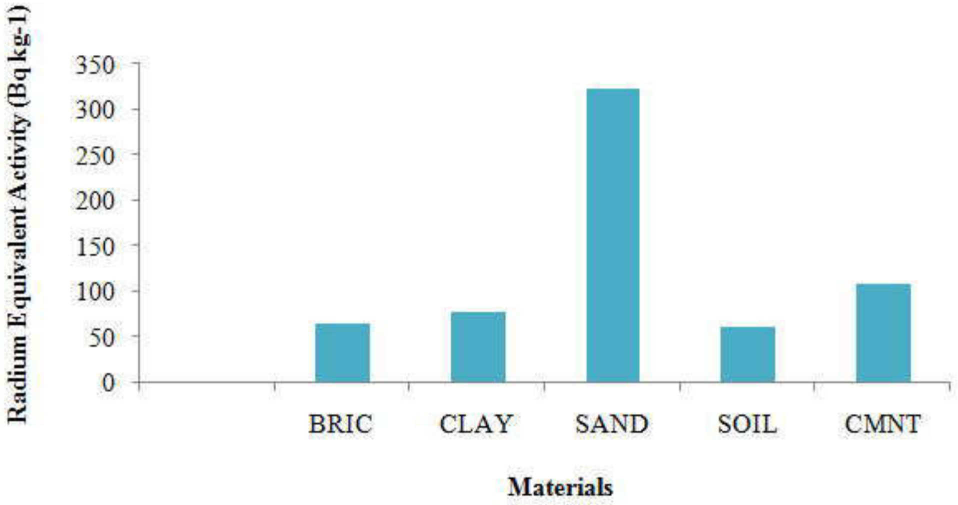

Figure 2 : Different types of Materials Vs Radium equivalent (Bq kg^{-1}).

The types of materials and its activity concentrations (Bq kg^{-1}) are shows in second, third and forth colum of Table 1, The fifth colum of Table 1 is shows the radium equivalent activities (Bq kg^{-1}) of different materials, The sixth colum of table 1 is shows the criteria formula ($mSvy^{-1}$) of different materials, The seventh colum of table 1 shows the absorbed does rates values ($nGyn^{-1}$) of different materials, The eighth and ninth columns of Table 1 is shows the annual effective dose rates values ($msvy^{-1}$) of different materials.

3.3. Criteria Formula

Based on models suggested by Krisiuk et al. (1971) and Stranden (1976), a value of 1.5 mGy was obtained by Krieger (1981) when evaluating the annual external radiation dose inside dwellings constructed of building materials with a Ra_{eq} value of

370 Bq kg^{-1}. These authors later corrected their calculations by taking into consideration a wall of finite thickness and applying a weighing factor of 0.7 (Keller and Muth, 1990) to account for the presence of windows and doors. Their results can be used as a criterion to limit the annual radiation dose from building materials based on the following formula:

$(A_{Ra} / 740 \text{ Bq/kg}) + (A_{Th} / 520 \text{ Bq/kg}) + (A_K / 9620 \text{ Bq/kg}) < 1$ --------------- (2)

where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in building materials in units of Bq kg^{-1}. The corresponding values calculated from the sum of the three quotients for the annual radiation dose associated with the studied building materials are given in the sixth column of Table-1.

The values range from 0.277 (Soil) to 1.27 (Sand) with an average of 0.09. The average value (0.1726) of the studied samples is below the recommended maximum value (<1). This indicates that gamma activity in the building materials does not exceed the proposed criterion level. Figure 3 shows the various types of building materials and the criterion formula.

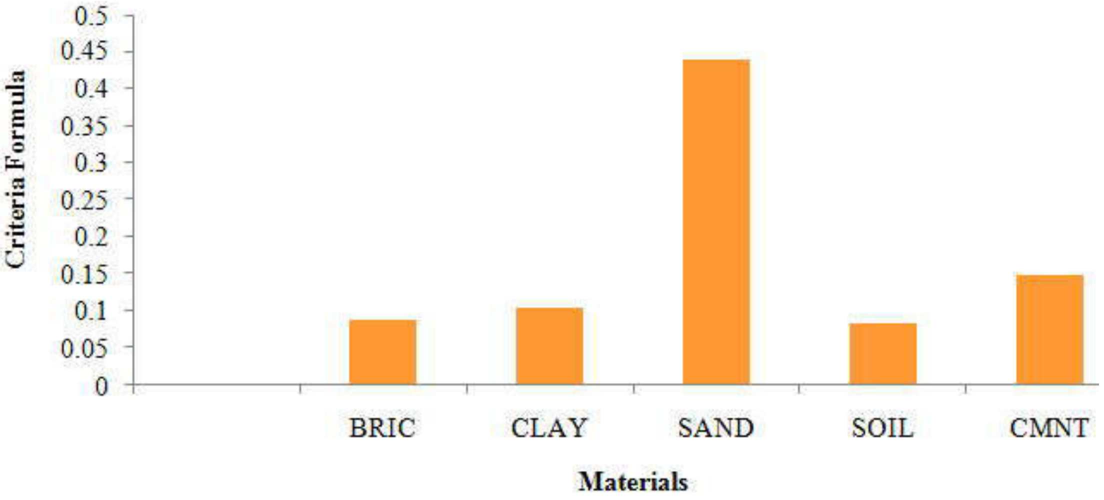

Figure 3 : Different types of Materials Vs Criteria Formula.

Estimation of the absorbed gamma dose rate (D_R) and the annual effective dose rate (H_R)

The absorbed dose rates in indoor air (D_R) and the corresponding annual effective doses (H_R) attributed to gamma-ray emission from the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials were evaluated using data and formulas provided by UNSCEAR (2000) and the EC (1999). In the UNSCEAR and European Commission reports, the dose conversion coefficients were calculated for the center of a standard room. The dimensions of this room are 4m×5m×2.8 m. The thickness of the walls, floors and ceiling and the density of the structure are 20 cm and 2350 kg m^{-3} (concrete), respectively. The resulting dose coefficients were found to be 0.92 nGy h^{-1} per Bq kg^{-1} for ^{226}Ra, 1.1 nGy h^{-1} per Bq kg^{-1} for ^{232}Th and 0.080 nGy h^{-1} per Bq kg^{-1} for ^{40}K.

$D_R(\text{nGy h}^{-1}) = 0.92 \times A_{Ra} + 1{:}1 \times A_{Th} + 0.080 \times A_K$ -------------------- (3)

where A_{Ra} is the activity concentration of ^{226}Ra, A_{Th} is the activity concentration of ^{232}Th, and A_K is the activity concentration of ^{40}K in units of Bq kg^{-1}.

To estimate the annual effective dose rates, it is necessary to use the conversion coefficient from the absorbed dose in air to the effective dose (0.7 $SvGy^{-1}$) and the outdoor occupancy factor (0.2 $SvGy^{-1}$) proposed by UNSCEAR (2000). Therefore, the effective dose rate is determined as follows:

Outdoor (mSv y^{-1}) = D_R (nGy h^{-1}) × 24 h × 365.25 d × 0.2 (out-door occupancy factor) × 0.7 Sv Gy^{-1} (conversion factor)×10^{-6}

$H_R = D_R \times 8766 \times 0{:}2 \times 0{:}7 \times 10^{-6} = D_R x 0{:}00123$ --------------------- (4)
where D_R (nGy h^{-1}) is given by Eq. (3).

The estimated results for D_R and the corresponding H_R values are given in the seventh and eighth columns of Table-1. The estimated D_R and H_R values for all the studied building materials range from 56.578 (Soil) to 259.256 (Sand) nGy h^{-1} and 0.277 (Soil) to 1.277 (Sand) mSv y^{-1}, respectively.

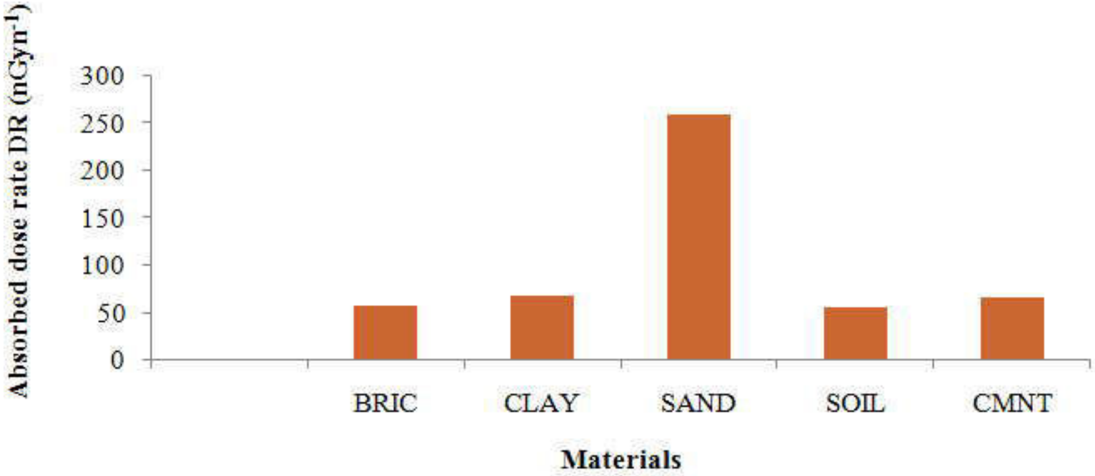

Figure 4 : Different types of Materials Vs Absorbed dose rate (nGy h^{-1})

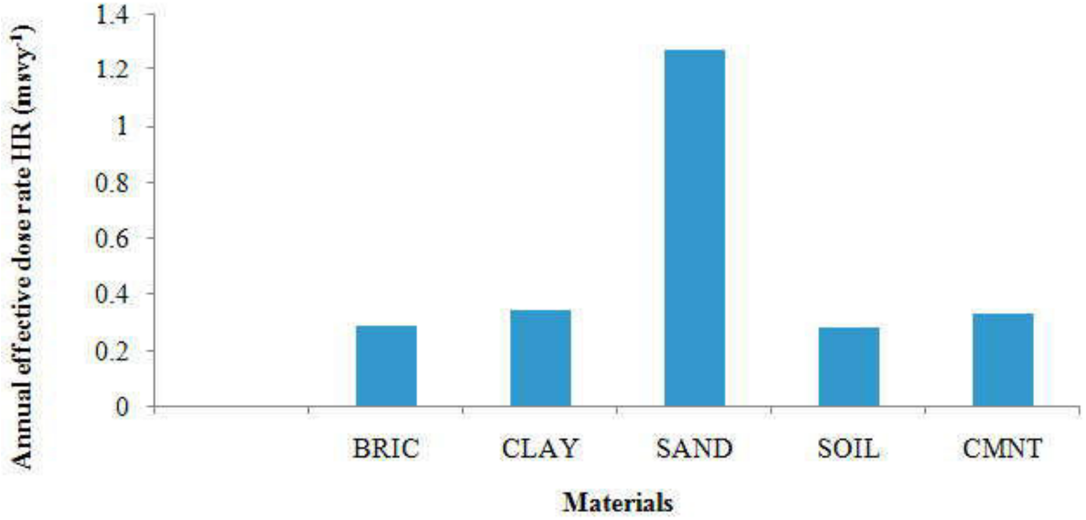

Figure 5 : Different types of Materials Vs Annual Effective dose rate (mSv y^{-1})

3.5. Radiation hazard indices

Beretka and Mathew (1985) defined two indices that represent (i) the external radiation hazard, H_{ex}, and (ii) the internal radiation hazard, H_{in}, which are discussed in this section. The tenth and eleventh colums of Table 1 is shows the external radiation hazards and internal radiation hazards.

3.5.1. External radiation hazard (H_{ex})

The external hazard index is an additional criterion to assess the radiological suitability of a material. It is defined as follows:

$H_{ex} = (A_{Ra} / 370\ Bq/kg) + (A_{Th} / 258\ Bq/kg) + (A_K / 4810\ Bq/kg)$ ----------(5)

where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in units of Bq kg^{-1}. The values of these indices should be less than unity for the radiation hazard to be negligible, i.e., for the radiation exposure attributed to radioactivity in construction materials to be limited to 1.50 mSv y^{-1}. Table 8 shows that the mean value, 0.34, of H_{ex} is below the criterion value (<1). Table 1 of tenth colum is indicate the external hazards (H_{ex}) of different materials and Figure 7 shows the various types of building materials and External radiation hazard H_{ex}.

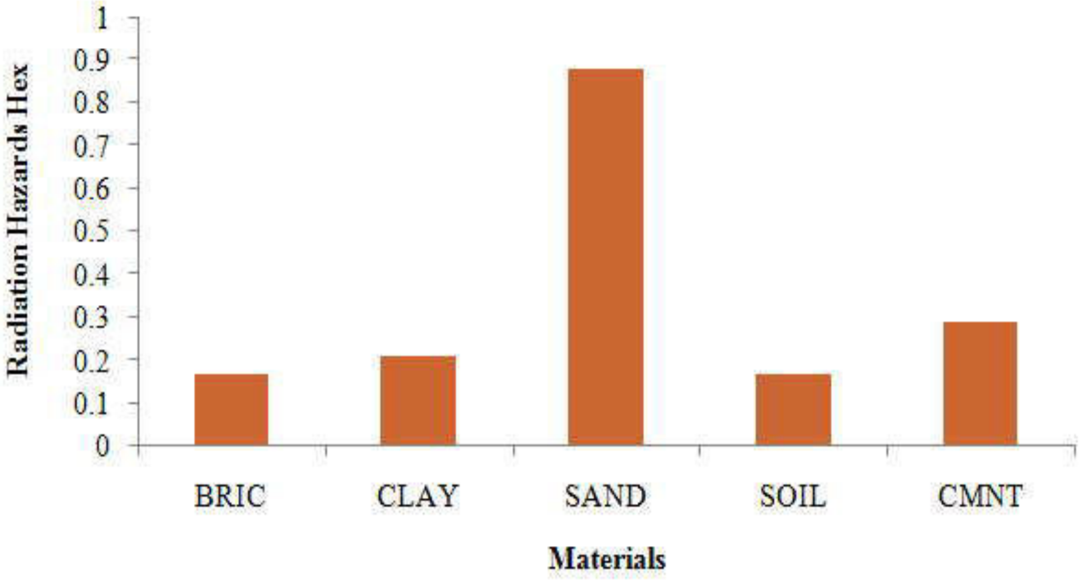

Figure 7 Different types of Materials Vs Radiation Hazards (H_{ex}).

3.5.2. Internal radiation hazard (H_{in})

In addition to the external radiation hazard they pose, radon and its short-lived daughters are also hazardous to the respiratory organs. The internal exposure caused by radon and its daughter products is quantified by the internal hazard index H_{in}, which has been defined as shown below:

$H_{in} = (A_{Ra} / 185 \text{ Bq/kg}) + (A_{Th} / 259 \text{ Bq/kg}) + (A_K / 4810 \text{ Bq/kg})$ ---------------- (6)

The internal hazard index is defined to reduce the acceptable maximum concentration of ^{226}Ra to half the value appropriate to external exposure alone. For the safe use of materials in the construction of dwellings, the following criterion was proposed by Krieger (1981):

$H_{in} \leq 1$ ----------------- (7)

The mean value of H_{in} is determined to be 0.37, which is <1, indicating that the internal hazard is below the critical value and it indicates that the materials are free from radiation hazards. Table 1 of eleventh colum is indicate the internal hazards (H_{ix}) of different materials, and Figure 8 shows the various types of building materials and internal radiation hazard $H_{in.}$

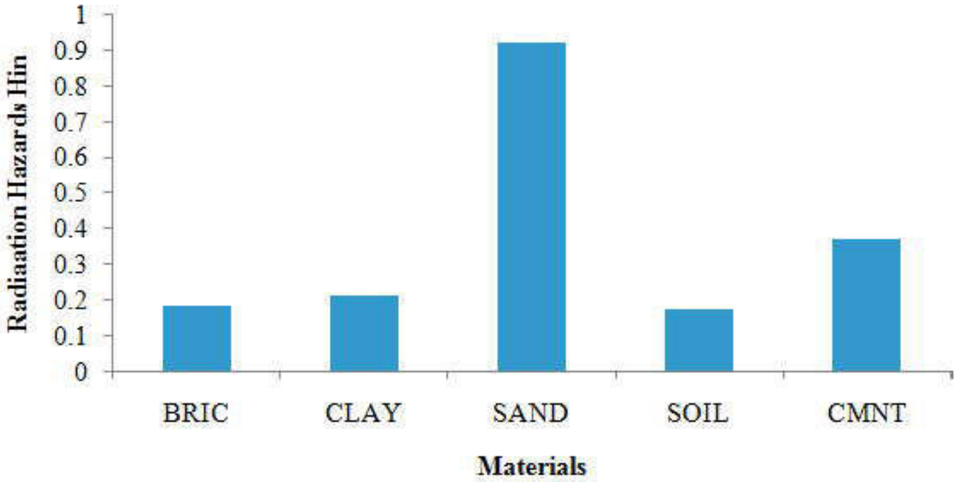

Figure 8 : Different types of Materials Vs Radiation Hazards (H_{in}).

4.0 Conclusion :

The specific radioactivity values of ^{226}Ra, ^{232}Th and ^{40}K measured in commonly used building materials used for the construction purposes in Sathanoor, Tiruvannamalai dist, Tamilnadu have been determined by gamma ray spectrometer. For each sample in this study, the specific activity, radium equivalent activity, criteria formula, the indoor gamma absorbed dose rate, annual effective dose rate and radiation hazard indices have been determined to assess the radiological hazards from the building materials.

The values obtained in the study are within the recommended safety limits, showing that these building materials do not pose any significant radiation hazard and hence the use of these materials in the construction for dwelling purpose can be considered to be safe for the inhabitants. This study can be used as a reference for more extensive studies of the same subject in future.

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