

Research Article

## FFA Study on Activity Concentration of Natural Radionuclides of Building Materials in Pachal, Tiruvannamalai dist, Tamilnadu, India

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Received Date: November 9, 2014

Accepted Date: January 27, 2015

Published Date: January 31, 2015

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**Citation:** Ravishankar, R., et al. FFA Study on Activity Concentration of Natural Radionuclides of Building Materials in Pachal, Tiruvannamalai dist, Tamilnadu, India. (2015) J Environ Health Sci 1(1): 1-5.

**Keywords:** Building Materials; Gamma Ray Spectrometry; Radiological Hazards

### Abstract

This paper presents the finding of a study undertaken to determine the natural radioactivity present in building materials in Pachal of Tiruvannamalai District, Tamilnadu. During this study, the samples of commonly used raw materials for buildings were collected and analyzed for primordial radionuclides by gamma ray spectrometry using NaI (TI) detector. The specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were measured in the samples. The radium-equivalent activity ( $Ra_{eq}$ ) of the samples was calculated and compared with similar data obtained in other countries that are reported in the literature. The comparison reveals that  $Ra_{eq}$  values obtained in our study area fall far below the criterion limit specified in building materials. The potential radiological hazard of the different samples was estimated using different approaches. These values were compared with the published data and were found to be within the acceptable limits. The results show that these materials are quite safe to be used as building materials.

### Introduction

Man is continuously exposed to ionizing radiation from naturally occurring radioactive materials. These ionizing radiations coming from building materials, air, water and food. Measurement of activity concentrations of radionuclides in building materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors. A knowledge of natural radioactivity in man and its environment is important since naturally occurring radionuclides are the major source of radiation exposure to man. An established fact that all the construction material contains trace amount of natural radioactivity. This activity in a major source of external and internal radiation exposure to the occupants of the dwelling. The natural radioactivity in soil and building materials come mainly from uranium (<sup>238</sup>U) and thorium (<sup>232</sup>Th) series and the radioactive isotope of potassium (<sup>40</sup>K). All these can be sources of both internal and external radiation exposure. Internal exposure occurs through inhalation of radon gas and external exposure occurs through the emission of penetrating gamma rays. Therefore, it is important to measure the radioactivity levels in the built-up areas to assess the radiological consequences. Even more important is the knowledge of the amount of natural activity present in the materials which are used in the construction of dwellings. The amount of activity present in building materials will decide its use in the construction of dwellings. The natural radioactivity of building materials in many countries has been reported<sup>[1-12]</sup>. Natural radioactivity in some Indian building materials has also been reported by other authors<sup>[6, 11-15]</sup>. A knowledge

of radioactivity present in construction materials helps to (a) assess the possible radiological hazards to human health and (b) develop the standards and guidelines for use and management of these materials. The present study was undertaken with the purpose of determining radioactivity in some building materials from Pachal of Tiruvannamalai District, Tamilnadu and assessing the radiological hazards due to external gamma ray exposure in dwellings.

### Methods of Measurements

**Sampling and Preparation:** Samples representing five different commonly used building materials were collected randomly from sites where housing and other buildings were under construction and from building material suppliers in Pachal Village for the measurement of the specific radioactivity of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K. Structural building materials (cement, brick, clay, sand and soil) are used in bulk amounts. The collected samples were kept in polyethylene bags which were numbered and catalogued for identification. The samples were brought to the sample preparation section of the low-level activity measurement laboratory. The brick samples were crushed, ground, and pulverised to a powder. The powder was passed through a sieve of 150 micron mesh size. The samples in powder form were dried at 110°C in a temperature-controlled furnace until there was no detectable change in the mass of the sample. The samples were transferred to radon-impermeable plastic containers of 6cm diameter and 6.5 cm height. Then these samples were sealed and the sealed containers were left for at least 4 weeks (>7 half-lives

of <sup>222</sup>Rn) before counting by gamma ray spectrometry in order to ensure that the daughter products of <sup>226</sup>Ra up to <sup>210</sup>Pb and <sup>228</sup>Th up to <sup>208</sup>Pb achieve equilibrium with their respective parent radio nuclides<sup>[11]</sup>. Their respective net weights were measured and recorded with a high sensitive balance.

**Gamma spectrometry analysis of samples**

A 3”x 3” NaI (TI) scintillation detector has been used for spectral measurements to enable one to cover the energy spectrum of the naturally occurring radio nuclides up to 2.6 MeV (<sup>208</sup>Tl, a daughter product of <sup>232</sup>Th). The detector is shielded by 15cm thick lead on all sides, including the top to reduce background due to cosmic ray component by almost 98%. The inner sides of the lead shielding is lined with 2mm thick Aluminium. Standard sources of the primordial radio nuclides obtained from IAEA in the same geometry and having the same density, as that of the prepared soil samples, were used to determine the efficiency of the detector for various energies in the prescribed geometry. The prepared samples were placed on top of the 3”x3” NaI (TI) detector and using the gamma ray spectrometer and multichannel analyzer, count spectra were obtained for each of the building material samples. The activity content of the three primordial radio nuclides viz, <sup>40</sup>K, <sup>232</sup>Th and <sup>226</sup>Ra are deduced from the count spectra. The region under the peaks corresponding to 1.46 MeV (<sup>40</sup>K), 1.764 MeV (<sup>214</sup>Bi) and 2.614 MeV (<sup>208</sup>Tl) energies are considered to arrive at the radioactivity levels of <sup>40</sup>K, <sup>226</sup>Ra and <sup>232</sup>Th, respectively. The minimum detectable activity (MDA) of each of the three primordial radio nuclides is determined from the background radiation spectrum obtained for the same counting time as was done for the samples and is estimated at 2.15 Bq/kg for <sup>232</sup>Th, 2.22 Bq/kg for <sup>238</sup>U and 8.83 Bq/kg for <sup>40</sup>K. All the building materials were subjected to gamma ray spectral analysis with a counting time of 20,000s.

**Results and Discussion**

**Radionuclide concentrations:** Table 1 showed that the average radionuclide concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and the radium equivalent activity beside the calculated criteria formula, total absorbed dose rate (D<sub>R</sub> in nGy/h), annual effective dose rate (in mSv/y) the external hazard index (H<sub>ex</sub>) and the internal hazard index (H<sub>in</sub>) for the investigated in the samples. It can be seen from the table 1 that the potassium isotope contributes more activity compared with other two isotopes. The lowest value of <sup>226</sup>Ra concentration is 7.47 Bq/kg measured in sand while the highest value is 31.26 Bq/kg measured in cement. The lowest value for <sup>232</sup>Th is 27.09 Bq/kg recorded in sand sample and the highest value is 77.8 Bq/kg obtained in clay sample. The lowest and highest values for <sup>40</sup>K were measured to be 233.92 Bq/kg and 419.7 Bq/kg in cement and soil respectively. The distribution of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in building materials is not uniform. To monitor the radioactivity levels of a new building and construction materials, different radiological indices of these materials should be determined.

**Table 1:** Activity concentration, Radium equivalent Activity (Ra<sub>eq</sub>), Criteria formula (CF) Absorbed Dose Rate (D<sub>R</sub>) Annual Effective Dose rate (H<sub>R</sub>), Internal and External radiation hazard indices (H<sub>in</sub> & H<sub>ex</sub> used in Pachal, Tiruvannamali Dist, Tamilnadu

S. No	Material	Activity concentration (Bq/kg)			Ra <sub>eq</sub> (Bq/kg)	Criteria formula (CF)	Absorbed Dose rate(D <sub>R</sub> ) (nGy/h)	Annual Effective Dose rate (H <sub>R</sub> ) (mSv/y)	H <sub>in</sub>	H <sub>ex</sub>
		<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K						
1	Brick	8.61	27.15	344.75	73.98	0.0997	65.36	0.08	0.22	0.199
2	Clay	BDL	77.8	414.5	143.17	0.1927	118.74	0.145	0.38	0.386
3	Soil	11.82	74.2	419.7	150.24	0.2023	126.07	0.154	0.43	0.405
4	Sand	7.47	27.09	293.21	68.79	0.0927	60.12	0.073	0.2	0.185
5	Cement	31.26	41.75	233.92	108.97	0.1468	93.39	0.114	0.37	0.294
AVERAGE		11.83	49.59	341.21	109.03	0.1468	92.74	0.113	0.32	0.294

**Radium Equivalent Activity (Ra<sub>eq</sub>)**

To represent the activity levels of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>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<sub>eq</sub>) activity and is mathematically defined as<sup>[3]</sup>

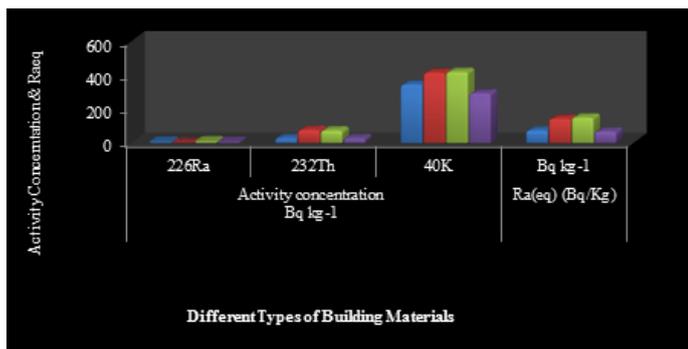
$$Ra_{eq} \text{ (Bq/kg)} = A_{Ra} + 1.43A_{Th} + 0.077A_K \text{ ----- (1)}$$

Where A<sub>Ra</sub>, A<sub>Th</sub>, A<sub>K</sub> represents the activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K. In the above relation, it has been assumed that 10 Bqkg<sup>-1</sup> of <sup>226</sup>Ra, 7 Bqkg<sup>-1</sup> of <sup>232</sup>Th and 130 Bqkg<sup>-1</sup> of <sup>40</sup>K produce equal γ- dose.

The recommended maximum levels of Ra<sub>eq</sub> for building materials to be used for homes and industries are < 370 Bq/kg. The calculated Ra<sub>eq</sub> values range from 68.79 (sand) to 150.24 (soil) Bq/kg with an average of 109.03 Bq/kg. All values of Ra<sub>eq</sub> in the studied samples are found to be lower than the criterion limit of 370 Bq/kg<sup>[16]</sup>. The results of this study show that the average value of Ra<sub>eq</sub> obtained for the building materials is 109.03 Bq/kg, which is below the recommended maximum value (370 Bq/kg) and thus do not pose any radiological hazard when used for the construction of buildings. The table 2 shows that the comparison of radium equivalents (Bq/kg) in clay bricks, cement, sand, soil/clay in different areas of the world. Fig-1 shows the building materials variation with activity concentration and radium equivalent activity

**Table 2.** Comparison of radium equivalents (Bq/kg) in clay bricks, Cement, Sand, Soil/Clay in different areas of the world.

S. No	Country	Clay bricks	Cement	Sand	Soil/ clay	Reference
1	Algeria	130	112	28	-	[2]
2	Brazil	202.7	188.8	102	68.2	[17]
3	Cameroon	193.34	70.1	104	-	[18]
4	China	201.9	127.7	94.7	-	[19]
5	Cuba	134.6	73.9	53.5	-	[20]
6	Jordan	-	-	86.22	-	[21]
7	NWFP	-	-	-	122	[22-23]
8	Bahawalpur division	-	-	-	158.5	[24]
9	Punjab	-	-	-	141	[25]
10	Paschal	73.98	108.97	68.79	150.24	Present work



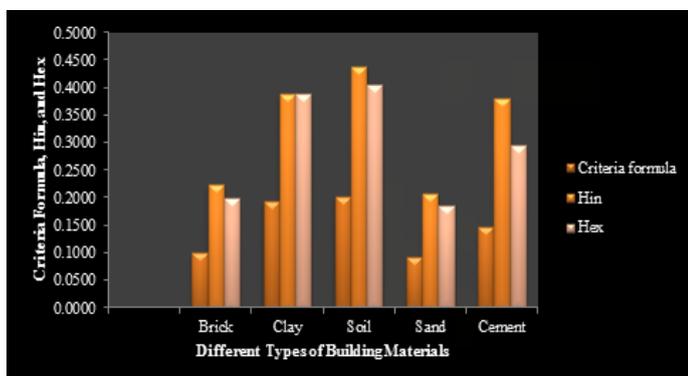
**Figure.1** Different types of Building Materials Vs Activity Concentration (Bq/kg) & Ra<sub>eq</sub> (Bq/kg)

**Criteria Formula (CF)**

Based on models suggested by Krisiuk et al<sup>[26]</sup> and Stranden<sup>[27]</sup>, a value of 1.5 mGy was obtained by Krieger<sup>[28]</sup> when evaluating the annual external radiation dose inside dwellings constructed of building materials with a Ra<sub>eq</sub> value of 370 Bq/kg. These authors later corrected their calculations by taking into consideration a wall of finite thickness and applying a weighing factor of 0.7<sup>[29]</sup> 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:

$$CF = \frac{A_{Ra}}{740} + \frac{A_{Th}}{520} + \frac{A_K}{9620} < 1 \quad \text{----- (2)}$$

Where A<sub>Ra</sub>, A<sub>Th</sub> and A<sub>K</sub> are the activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K, respectively in building materials in units of Bq/kg. 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 Table-1. The values of criteria formula are ranging from 0.0927 (sand) to 0.2023 (soil) with an average of 0.1468. The average value (0.1468) 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 and safely used for constructions. Fig-2 shows the various types of building materials and the criteria formula.



**Figure 2:** Different types of Building Materials Vs Criteria formula, H<sub>in</sub> and H<sub>ex</sub>

**Estimation of Absorbed Gamma Dose Rate (D<sub>R</sub>) and Annual Effective Dose Rate (H<sub>R</sub>)**

The absorbed dose rates in indoor air (D<sub>R</sub>) and the corresponding annual effective doses (H<sub>R</sub>) attributed to gamma-ray emission from the radio nuclides (<sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K) in building materials were evaluated using data and formulae provided

by UNSCEAR<sup>[30]</sup> and the EC<sup>[31]</sup>. 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<sup>-3</sup> (concrete), respectively. The resulting dose coefficients were found to be 0.92 nGy h<sup>-1</sup> per Bq/kg for <sup>226</sup>Ra, 1.1 nGy h<sup>-1</sup> per Bq/kg for <sup>232</sup>Th and 0.080 nGy h<sup>-1</sup> per Bq/kg for <sup>40</sup>K.

$$D_R \text{ (nGyh}^{-1}\text{)} = 0.92 \times A_{Ra} + 1.1 \times A_{Th} + 0.080 \times A_K \text{ ----- (3)}$$

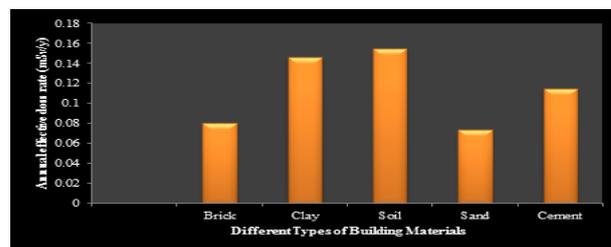
Where A<sub>Ra</sub> is the activity concentration of <sup>226</sup>Ra, A<sub>Th</sub> is the activity concentration of <sup>232</sup>Th, and A<sub>K</sub> is the activity concentration of <sup>40</sup>K in units of Bq/kg.

To estimate the annual effective dose rate, it is necessary to use the conversion coefficient from the absorbed dose in air to the effective dose (0.7 Sv Gy<sup>-1</sup>) and the outdoor occupancy factor (0.2 Sv Gy<sup>-1</sup>) proposed by UNSCEAR<sup>[30]</sup>. Therefore, the effective dose rate is determined as follows:

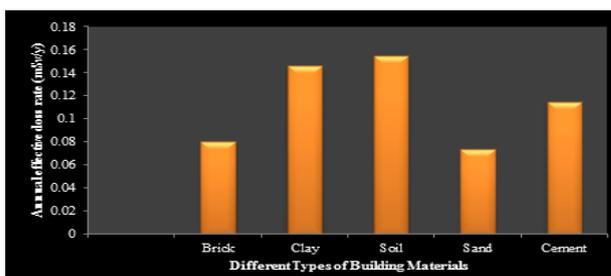
$$\text{Outdoor (mSv y}^{-1}\text{)} = D_R \text{ (nGy h}^{-1}\text{)} \times 24 \text{ h} \times 365.25 \text{ d} \times 0.2 \text{ (outdoor occupancy factor)} \times 0.7 \text{ Sv Gy}^{-1} \text{ (conversion factor)} \times 10^{-6}$$

$$H_R = D_R \times 8766 \times 0.2 \times 0.7 \times 10^{-6} = D_R \text{ 00123 ----- (4)}$$

Where D<sub>R</sub> (nGy h<sup>-1</sup>) is given by Eq. (3). The estimated result for D<sub>R</sub> and for H<sub>R</sub> is given in Table 1. The estimated D<sub>R</sub> and H<sub>R</sub> values for all the studied building materials range from 60.128 (sand) to 126.070 (soil) in nGy h<sup>-1</sup> and 0.0737 (Sand) to 0.1546 (soil) in mSv y<sup>-1</sup> respectively. From the data in Table-1, the estimated mean value of D<sub>R</sub> in the studied samples is 92.74 nGy h<sup>-1</sup>, which is slightly higher than world average (populated-weighted) indoor absorbed gamma dose rate of 84 nGy h<sup>-1</sup>. Additionally, the estimated mean value of the annual effective dose rate of 0.1137 mSv y<sup>-1</sup> is less than the permissible limit. Figs. 3 & 4 shows that the various types of building materials along with the absorbed dose rate and the annual effective dose rate respectively.



**Figure 3:** Different types of Building Materials Vs Absorbed Doss Rate (nGy/h)



**Figure 4:** Different types of Building Materials Vs Annual effective doss rate (mSv/y)

### Radiation hazard indices

Beretka and Mathew<sup>[3]</sup> defined the two indices that represent (i) the internal radiation hazard,  $H_{in}$  (ii) the external radiation hazard,  $H_{ex}$ , which are discussed in this section.

#### Internal Radiation hazard index ( $H_{in}$ )

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:

$$H_{in} = \frac{A_{Ra}}{185Bq/kg} + \frac{A_{Th}}{259Bq/kg} + \frac{A_K}{4810Bq/kg} \text{ -----(5)}$$

The internal hazard index is defined to reduce the acceptable maximum concentration of  $^{226}\text{Ra}$  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<sup>[28]</sup>

$$H_{in} \leq 1 \text{ ---- (6)}$$

The mean value of  $H_{in}$  is determined to be 0.32 which is  $<1$ , indicating that the internal hazard is below the critical value and it indicates that the materials are free from radiation hazards. Fig-2 shows the various types of building materials and internal radiation hazard  $H_{in}$ .

#### External Radiation hazard index ( $H_{ex}$ )

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

$$H_{ex} = \frac{A_{Ra}}{370Bq/kg} + \frac{A_{Th}}{259Bq/kg} + \frac{A_K}{4810Bq/kg} \leq 1 \text{ -----(7)}$$

Where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively in units of Bq/kg. The value of this index should be less than unity for the radiation hazard is to be negligible, i.e., for the radiation exposure attributed to radioactivity in construction materials to be limited to  $1.50 \text{ mSv y}^{-1}$ . The mean value 0.29 of  $H_{ex}$  is below the criterion value ( $<1$ ) indicates that no significant radiation hazards of building materials. Fig-2 shows the various types of building materials and External radiation hazard index  $H_{ex}$ .

### Conclusion

The natural radionuclide activity content, radium equivalent activity, the criteria formula, the absorbed gamma dose rate in indoor air and the corresponding annual effective dose, the external and internal hazard indices of some building materials commonly used in Pachal village of Tiruvannamalai district, Tamilnadu, India, were determined. All the health hazard indices are well below their recommended limits. The building materials investigated in this study can be safely used as building material for dwelling construction. The obtained data could be useful as base line data for radiation assessment in building materials.

### Acknowledgment

One of the author (R. Ravisankar) wishes to express his high

gratitude to Dr. B. Venkatraman, AD, RSEG, Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam, Tamilnadu for giving his permission to use the nuclear counting facility in RSD and also Mr. R. Mathiarasu, Scientific Officer, RSD, IGCAR, Kalpakkam, India for his technical help in counting the samples. Our special thanks to Dr. M.T. Jose, Head, RSD, IGCAR for his keen help, constant encouragements in Gamma ray spectroscopic measurements.

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