

Radiometric analysis of some building materials using gamma‑ray spectrometry

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Abstract

The present study focuses on the efect of natural radioactivity of some common building materials to humans. The main radiological risk assessment parameters such as radium equivalent, dose rate, annual efective dose equivalent and hazard indices were determined using Gamma ray spectrometry for individual building materials and their three diferent combinations. On comparing the results with world average values, it is inferred that the building materials do not pose any radiation hazards. Also as all the radiological parameters for M-sand are much lesser than that of river sand, it can be a good replacement for river sand.

Keyword Building materials · Gamma ray spectrometry · Radionuclides · Radiation hazards

Introduction

Humans are continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM). The origin of these radioactive materials is found in the earth's crust [[1\]](#page-7-0). They fnd their way into building materials through construction activities [\[2\]](#page-8-0). All building materials contain diferent amounts of radioactive substances which originate from rocks and soil [[3](#page-8-1)]. They contain natural radionuclides of the 238U, 232Th series and the radioactive isotope of potassium $({}^{40}K)$ [[4\]](#page-8-2). In the uranium series, the decay chain segment starts from ²²⁶Ra which is the most important one and therefore reference is often made to 226 Ra instead of 238 U [\[5,](#page-8-3) [6](#page-8-4)]. Also 226 Ra and 232 Th increase the concentration of 222 Rn and 220 Rn and their daughter products in buildings. The inhalation of Rn isotopes and their short-lived radionuclides leads to internal exposure and is considered as the major source of radiation disease [[7,](#page-8-5) [8](#page-8-6)]. External exposure occurs through the emission of penetrating gamma rays [\[9](#page-8-7)]. Hence, natural radioactivity is a major source of external and internal exposure to the humans in dwellings. Longterm exposure to uranium and radium through inhalation has several health effects as chronic lung diseases, acute

 \boxtimes V. Thangam thangamj3@gmail.com leucopoenia, anemia and necrosis of the mouth. Radium causes bone, cranial, and nasal tumors. Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukemia [[10](#page-8-8)]. Measurement of activity concentrations of radionuclides in building materials are of utmost importance in order to assess the radiological risk to human health [\[11](#page-8-9)]. The world average activity concentrations of $226Ra$, $232Th$ and 40 K in building materials are 50 Bqkg⁻¹, 50Bqkg⁻¹ and 500 Bqkg⁻¹ respectively $[12]$ $[12]$.

In recent days, manufactured sand (M-Sand) is used as a substitute for river sand in concrete construction. It is manufactured from rock deposits (hard granite rocks) by squashing. This crushed sand is of cubical shape with grounded edges, washed and graded to as a construction material. M-Sand presents various favorable advantages including its contribution as fller content and in diminishing environmental impact $[13–15]$ $[13–15]$ $[13–15]$. Also, M-sand has the advantage of being free of impurities such as dust, clay and silt. This avoids impairing of bond between the cement paste and the aggregates by reducing the water requirement unlike in river sand. Also size can be controlled as required for any given construction. All the above increase the quality and durability of construction.

In the present study, diferent types of basic building materials like river sand, cement, M-sand, red brick, white cement and limestone were collected from Tirunelveli district, Tamilnadu, India. The district has a geographical area of 6759 sq.km and lies in the south eastern part of

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Tamilnadu state and is bounded by the coordinates 08°05′ to 09°30′N and 77°05′ to 78°25′E. The perennial river Tamirabarani fows almost in the central part of the district and feeds the entire district population. The river is located between the geographic co-ordinates 8° 26′ 45" to 9° 12′ 00"N and 77° 09′ 00" to 78° 08′ 30"E. The length is 120 km and the area is about 5717.08 sq km. The origin of this river is beside the mountain slopes of the Western Ghats leading to the south-west Malabar Coast (Kerala coast) that is also noted for its high background radiation [[16\]](#page-8-13). Hence this work was taken up.

The main objective of the present study was to determine the activity concentrations of the three radionuclides 226 Ra, 232 Th and 40 K in several types of building materials used in Tirunelveli district, Tamilnadu, India from natural resources. The results of these studies were compared with the results of similar studies of other countries in the world. Also, since any construction is a combination of the basic building materials, three diferent combinations like combination-1 (river sand (RS), cement (CT), red brick (RB), white cement (WC) and limestone (LS)), combination-2 (manufactured sand (MS), CT, RB, WC and LS) and combination-3 (RS, MS, CT, RB, WC and LS) (overall) were analyzed. The composition of combinations 1, 2 and 3 are chosen with an aim to fnd if it is advantageous radilogically to replace river sand with M-sand. Hence keeping other basic building materials such as cement, red brick, white cement and limestone same, M-sand is used in combination 1, river sand is used in combination 2 and a mixture of river and M-sand is used in combination 3. Equal proportion of all the building materials are used for mixing in all the three combinations.

Experimental analysis

Sample collection and preparation

In the present study, diferent types of most commonly used building materials in Tirunelveli district Tamilnadu, India were collected from housing sites, manufacturers and suppliers. River sand was collected from ten diferent locations of Thamirabarani river with the distance between the sampling sites being around 5 km.

The collected samples were crushed into a fne powder using a grinder with a grain size of 1 mm and heated at 130 °C in the hot air oven until the sample weight became constant to get rid of humidity and moisture if any. The prepared samples were stored into radon impermeable plastic containers and were sealed by cellophane tapes. The packed samples were kept aside for 30–40 days to bring 222 Rn and its short-lived daughter products into equilibrium with 226Ra.

Radiometric analysis

Activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for the building materials were measured using a Gamma ray spectrometer with a $3" \times 3"$ NaI (Tl) based detector. Efficiency calibration of the detector was accomplished using International Atomic Energy Agency (IAEA) standard reference materials such as RG-U, RG-Th and RG-K. Both background and samples were counted for a period of 20,000 s. The gamma ray photo peaks corresponding to 1460 keV for $40K$, 1764 keV for ²¹⁴Bi and 2614 keV for 208Tl were used for determining the activity concentrations of 40 K, 238 U and 232 Th respectively. Minimum Detectable Activity (MDA) or Below Detectable Limit (BDL) of the NaI (Tl) system for the three nuclides ²³⁸U, ²³²Th and ⁴⁰K are 7 Bq kg⁻¹, 8 Bq kg⁻¹ and 30 Bq kg⁻¹ respectively. The samples were then placed on the top of the detector and the spectra was recorded for 20,000 s.

Results and discussion

The calculated values of activity concentrations of the radionuclides ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K, radium equivalent activity (Ra_{eq}), dose rate (D_R), annual effective dose equivalent (AEDE), hazard indices and activity utilization index for diferent types of building materials and their combinations are reported in Tables [1,](#page-2-0) [2](#page-3-0) and [3](#page-4-0).

Analysis of radiological parameters

Activity concentration

Activity concentrations of three radionuclides ²²⁶Ra $(2^{38}U)$, $2^{32}Th$ and $40K$ were estimated in different types of building materials and are listed in Table [1](#page-2-0). The activity concentration of 226 Ra varies from BDL (river sand) to 192 ± 15 Bqkg⁻¹ (cement). The activity of ²³²Th ranged from BDL (M-sand) to 127 ± 14 Bqkg⁻¹ (river sand). The activity concentration of ⁴⁰K varies from 192 ± 60 Bqkg⁻¹ (cement) to 1572 ± 65 Bqkg⁻¹ (M-sand).

The mean values of activity concentrations for different type of building materials are also listed in Table[1](#page-2-0) and depicted in Fig. [1.](#page-4-1) It can be easily seen from the figure that the value of activity concentrations of the radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K follows the pattern $LS > CT > WC > RB > RS > MS$, $RS > RB > CT > WC > LS > MS$ and $LS > RS > MS > RB > WC > CT$. The maximum values are observed for ²²⁶Ra, ²³²Th and ⁴⁰K in LS, RB and LS

Table 1 Activity concentrations of 226 Ra, 232 Th and 40 K and Ra_{eq} in Building materials from Tirunelveli district, Tamilnadu, India

 $(160 \pm 15, 53 \pm 15 \text{ and } 1017 \pm 70 \text{ Bqkg}^{-1} \text{ respectively}).$ The lowest values are observed for the same nuclides in MS, MS and CT (36 \pm 10, BDL, 294 \pm 59 Bqkg⁻¹).

The mean activity concentrations for combinations 1, 2 and 3 are given in Table [3](#page-4-0), and depicted in Fig. [5](#page-6-0). It can be observed that the value of activity concentrations of ^{226}Ra , ²³²Th and ⁴⁰K follows the pattern Combination 1 > Combination $2 >$ Combination 3; Combination $1 >$ Combination 3>Combination 2 and Combination 3>Combination 1>Combination 2.

The mean values of activity concentrations of ^{226}Ra , ^{232}Th and 40K for the basic building materials were compared with similar studies in other countries and with world average values and is shown in Table 4 [2 , 1 2 , 1 7 – 20].

Radium equivalent activity (Raeq)

The distribution of the natural radionuclides in all samples were found to be non-uniform. The uniformity with respect to the exposure of radiation can be defned in terms of radium equivalent activity (Ra_{eq}) [\[21](#page-8-16)]. It can be estimated for all samples in Bqkg−1 and calculated by the following formula (Eq. [1](#page-3-1)) [\[22](#page-8-17)],

Table 2 Gamma dose rate, annual efective dose equivalent and hazard indices of building materials

$$
Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K
$$
 (1)

where A_{Ra} , A_{Th} , A_{K} are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K. It is assumed that 10 Bqkg⁻¹ of ²²⁶Ra, 7 Bqkg⁻¹ of ²³²Th and 130 Bqkg⁻¹ of ⁴⁰K produce an equal gamma ray dose.

The sixth column of Table [1](#page-2-0) gives the Ra_{eq} of the building materials along with the mean for each type. Figure [1](#page-4-1) shows the variation of mean values of Ra_{eq} for each type of building material. The calculated values of Ra_{eq} of all building materials ranged from 33.66 Bqkg⁻¹ (M-sand) to 318.65 Bqkg⁻¹ (river sand). The mean values of Ra_{eq} follows the pattern $LS > CT > RB > RS > WC > MS$. Average values of Ra_{eq} for

 $Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K$ (1) combinations 1, 2 and [3](#page-4-0) are listed in Table 3 and also shown in Fig. [5](#page-6-0). It can be deciphered that the Ra_{eq} values are in the order Combination 1>Combination 2>Combination 3. All values are much lesser than the world average value (370 $Bqkg^{-1}$) [\[23\]](#page-8-18).

Dose rate (D_R)

Determination of Dose rate (D_R) is the most important aspect for evaluating the radiation exposure to the gamma radiation. It represents the mean energy imparted to matter per unit mass by the ionizing radiation. Dose rate can be calculated by the formula (Eq. [2\)](#page-4-2) [[24](#page-8-19)],

Table 3 Radiological parameters for three combinations of building materials

district, Tamilnadu

Fig. 1 Mean values of activity concentrations and Ra_{eq} of building materials in Tirunelveli

$$
D_{\rm R} = 0.462 A_{\rm Ra} + 0.604 A_{\rm E} + 0.0417 A_{\rm K}
$$
 (2)

^D^R ⁼ 0.462 *^A*Ra ⁺ 0.604 *^A*^E ⁺ 0.0417 *^A*^K (2) **Annual efective dose equivalent (AEDE)**

where A_{Ra} , A_{Th} , A_K are the activity concentrations of ²²⁶Ra, 232 Th, 40 K and 0.462, 0.604, 0.042 are the conversion factors used to convert the activity concentration to absorbed dose rate (D_R) in air per unit activity concentration in Bq kg−1. The computed values of absorbed dose rate for all the studied samples vary from 17.10 nGyh−1 (M-sand) to 145.97 $nGyh^{-1}$ (river sand) as seen from Table [2](#page-3-0). The mean values are also given in Table [2](#page-3-0) and shown in Fig. [2](#page-5-1). They follow the sequence LS>CT>RS>RB>WC>MS. Average D_R values for combinations 1, 2 and [3](#page-4-0) are listed in Table 3 and illustrated in Fig. [5.](#page-6-0) It follows the pattern Combination 1>Combination 2>Combination 3. The 1st combination using river sand has a value slightly higher than the popula-tion-weighted average of 84 nGyh⁻¹ [[23\]](#page-8-18).

Annual efective dose equivalent for diferent types of building materials is given in Table [2](#page-3-0) along with the mean values and is shown in Fig. [3.](#page-5-2) It was calculated by the conversion factor 0.7 SvGy−1 which was used to convert the absorbed dose to human effective dose equivalent with an indoor and outdoor occupancy of 80% and 20% respectively. AEDE was determined by the following formula (Eq. [3\)](#page-4-3) [\[6](#page-8-4)],

$$
AEDE_{\text{indoor}}(mSvy^{-1}) = D_R(nGyh^{-1}) \times 8760 h
$$

× 0.8 × 0.7SvGy⁻¹ × 10⁻⁶ (3)

The estimated values of indoor annual effective dose rate was least for M-sand (0.08 mSvy^{-1}) and highest for river

Table 4 Comparison of activity concentrations of building materials in Tirunelveli district with the results of similar studies reported in diferent countries

Fig. 2 Mean values of absorbed dose rate of building materials

Fig. 3 Mean values of annual effective dose equivalent for building materials

sand (0.72 mSvy⁻¹). The mean value of each type followed the pattern LS>CT>RS>RB>WC>MS.

The mean AEDE values for combinations 1, 2 and 3 are given in Table [3](#page-4-0) and depicted in Fig. [6](#page-7-1). It can be observed that the values of AEDE are in the order Combination 1>Combination 2>Combination3. The above result reveals that AEDE values of all the building materials are within the recommended limit (1 mSvy^{-1}) [\[23](#page-8-18)].

Hazard indices

To measure the hazards one can defne radiation hazard indices like external (H_{ex}) and internal (H_{in}) hazard indices which are calculated by the following relations (Eq. [4\)](#page-5-3) and (Eq. [5\)](#page-5-4) [[24](#page-8-19)],

$$
H_{\rm ex} = \frac{A_{\rm Ra}}{370} + \frac{A_{\rm Th}}{259} + \frac{A_{\rm K}}{4810}
$$
(4)

The internal exposure to 222 Rn and its daughter products can be assessed by the internal hazard index H_{in} [\[25](#page-8-20)].

$$
H_{\rm in} = \frac{A_{\rm Ra}}{185} + \frac{A_{\rm Th}}{259} + \frac{A_{\rm K}}{4810}
$$
 (5)

where H_{ex} , H_{in} are external and internal hazard indices. The estimated values of H_{ex} and H_{in} was least for M-sand (0.09 and 0.13). The calculated values of H_{ex} and

Fig. 4 Mean values of H_{ex} , H_{in} , I_{y} and AUI of building materials

 H_{in} were highest for river sand (0.86) and cement (1.28) (Table [2](#page-3-0) and Fig. [4\)](#page-6-1). It can be easily seen that the mean values of H_{ex} and H_{in} of all investigated materials follow the pattern $LS > CT > WC > RS > RB > MS$ and LS>CT>RB>WC>RS>MS respectively. The maximum average values are observed for H_{ex} and H_{in} in LS (0.69 and 1.12). The lowest values are observed for the same in MS (0.23 and 0.33) (Fig. [5](#page-6-0)). It can be observed that the value of H_{ex} and H_{in} follow the pattern Combination 1 > Combination 2>Combination3 and Combination 1>Combination 2>Combination 3 which is given in Table [3](#page-4-0) and depicted in Fig. [6](#page-7-1). Average values of H_{ex} and H_{in} in all the studied samples are within the world average value (≤ 1) [[23\]](#page-8-18).

Gamma index (*I***γ)**

Activity concentration index, or gamma index (I_v) must be less than 1 if the material is to be used in construction of interiors [\[12](#page-8-10)]. It was estimated by the following formula (Eq. [6](#page-6-2)) [\[26\]](#page-8-22),

$$
I_{\gamma} = \frac{A_{\text{Ra}}}{300} + \frac{A_{\text{Th}}}{200} + \frac{A_{\text{K}}}{3000}
$$
 (6)

where A_{Ra} , A_{Th} , A_K are the activity of concentrations ²²⁶Ra, 232 Th and 40 K respectively. The calculated values of gamma index was least for M-sand (0.13) and highest for river sand (1.16) (Table [2\)](#page-3-0). The mean values are also given in Table [2](#page-3-0) and the same is shown in Fig. [4](#page-6-1). The variation is of the form $LS > CT > RS > RB > WC > MS$. Combinations 1, 2 and 3 are listed in Table. [3](#page-4-0) and depicted Fig. [6](#page-7-1). It can be inferred that Combination $1 >$ Combination $2 >$ Combination 3. These values are less than the recommended limit (≤ 1) [\[23](#page-8-18)].

Activity utilization index (AUI)

Activity utilization index was used to investigate whether the samples can be used as building materials and was calculated by the following formula (Eq. [7](#page-6-3)) [\[27\]](#page-8-23),

$$
AUI = \frac{A_{Ra}}{50} f_{Ra} + \frac{A_{Th}}{50} f_{Th} + \frac{A_K}{50} f_K
$$
 (7)

Fig. 6 Mean values of AEDE, H_{ex} , H_{in} , I_{γ} and AUI for combinations 1, 2 and 3

where A_{Ra} , A_{Th} , A_K are the activity of ²²⁶Ra, ²³²Th, ⁴⁰K and $f_{R_0}(0.462)$, $f_{Th}(0.604)$, $f_K(0.041)$ are the fractional contributions to the total dose rate in air due to $γ$ -radiation from the actual activity concentrations of 226 Ra, 232 Th, 40 K respectively [\[28\]](#page-8-24). The computed values of AUI was least for M-sand (0.16) and highest for cement (2.46) (Table [2](#page-3-0)). The mean values are also shown in Fig. [4.](#page-6-1) It can be easily seen from the fgure that the value of AUI of all investigated materials follows the pattern $CT > LS > RB > WC > RS > MS$. Also, the values follow the order Combination $1 >$ Combination 2>Combination3 which is given in Table [3](#page-4-0) and depicted in Fig. [6](#page-7-1). Average values of AUI are within the world average value (≤ 2) [[23\]](#page-8-18).

Conclusion

In the present study, some basic building materials like river sand, M-sand, cement, red brick, white cement, limestone were subjected to gamma ray spectrometric analysis. Activity concentrations of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K were determined. Other radiological parameters such as R_{eq} , D_R , AEDE, H_{ex} , H_{in} , I_{γ} and AUI for all the studied samples were also calculated. These were compared with the world average values which is given by United Nations Scientifc Committee on the Efects of Atomic Radiation (UNSCEAR 2000). It can be inferred that mean values of all the radiological parameters except the activity concentration of 40 K are least for M-sand and well within the world average value. But as already stated in the introduction, 40 K does not lead to inhalation of Rn isotopes and their short-lived radionuclides and hence is not a source of major concern for internal exposure. Also from Tables [1](#page-2-0) and [2](#page-3-0), it can be easily seen that all the mean value of all parameters for river sand are higher than that for M-sand with some values being almost double. Thus, M-sand can be a very good substitute for river sand in construction activities.

Also three diferent combinations like combination-1, 2 and 3 were characterized for all materials. These results indicate that the most important risk assessment factors like Ra_{eq} , D_R , AEDE and H_{in} are least in combination 3 (overall). Combination 2 which contains M-sand has marginally higher values and is lesser in comparison to the combination 1 which contains river sand. Hence M-sand can be a good replacement for river sand. The mean values of the above parameters for the individual basic building materials are also well within the world average values except for the D_R values of cement and limestone which are slightly higher. However, since only a combination of these building materials go into construction, an analysis of the three combinations cited above shows that all the combinations have lower values than the world average values for all the radiological parameters barring the D_R value in combination 1. Out of the three combinations, the radiological parameters in the third combination are the lowest. Hence the third combination seems to be the best one followed by the second one for construction activities. However, as already stated, while considering the three combinations, only equal proportions of all building materials were used for mixing. A more comprehensive study by changing the proportions of diferent building materials in each combination is required as it would have a significant effect on the difference in radioactivity in each combination samples. This will form part of our next major study to analyze the efect of mixing ratios on radioactivity.

Even so, from the above arguments, it can be concluded that these materials do not pose any hazards to dwellers in buildings. Further work is in progress for more building materials like tiles, granite, marble, hollow bricks, fy-ash bricks etc. which form part of today's modern buildings. Also, statistical analysis is being done to analyze the correlation among various radiological parameters.

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