

MEASUREMENT OF NATURAL RADIOACTIVITY OF BUILDING MATERIALS USED IN THE THOUBAL DISTRICT OF MANIPUR, INDIA

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Abstract

A study of natural radionuclides of different types of houses was conducted at 222 houses in the Thoubal district of Manipur, India. The average annual effective doses of radiation (gamma) in indoor and outdoor were obtained from the measurements using Micro R-survey meter as 1.23 ± 0.10 (range: 1.02 - 1.35) mSvy⁻¹ and 0.76 ± 0.08 (range: 0.60 - 0.89) mSvy⁻¹ for reinforced cement concrete (RCC) houses, followed by 1.09 ± 0.10 (range: 0.95 - 1.19)mSvy⁻¹ and 0.75 ± 0.08 (range: 0.67 - 0.95) mSvy⁻¹ for Adobe laid (AL) earthen houses, 1.03 ± 0.09 (range: 0.78 - 1.24) mSvy⁻¹ and 0.74 ± 0.08 (range: 0.65 - 0.91) mSvy⁻¹ for Assam-type (AT) brick houses, 0.82 ± 0.09 (range: 0.64 - 0.99) mSvy⁻¹ and 0.72 ± 0.08 (range: 0.59 - 0.94) mSvy⁻¹ for AT mud houses and 0.70 ± 0.08 (range: 0.66 - 0.79) mSvy⁻¹ and 0.70 ± 0.07 (range: 0.65 - 0.82) mSvy⁻¹ for AT katcha houses. The activity concentration (²²⁶Ra, ²³²Th and ⁴⁰K) of building materials available in this district were evaluated using gamma ray spectrometry and found average value as 43.0 (range: 24.1 - 70.2) Bqkg⁻¹, 108.6 (range: 21.3 - 211.8) Bqkg⁻¹ and 1487.2(range: 998.7 - 2298.6) Bqkg⁻¹. Moreover, the annual effective dose conceived from building material was found as 0.7 mSvy^{-1} from Portland cement, 1.0 mSvy^{-1} each both for sand and brick.

Key words: Natural radioactivity, building materials, Annual effective dose rate, Thoubal district

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INTRODUCTION

radionuclides Natural are found in our knowledge environment and the of this radionuclides concentration level and their distribution in the environment has become a focus of much attention in assessing the human risk from natural radiation exposure. Most of the general population spends their maximum time (about 80%) in indoors, and there is a high chance of getting both external as well as internal radiation exposure from natural radionuclide present in the building materials [1-3]. Most of building materials used for constructing our houses such as concrete, brick, sand, marble, granite, limestone, gypsum etc mainly contain natural radionuclides such as Uranium (²³⁸U) and Thorium (²³²Th) and their decay radionuclide products and radioactive Potassium (40 K) [4]. In 238 U series, 98.5% of the radiological effects are produced by radium and its daughter radionuclide products, so radium and its decay chain segment is radiologically very important [4]. Radon, thoron and their progenies contribute maximum radiation dose to the occupational workers and general public [5]. These two radionuclide gases present in the ambient air as well as in the indoor atmosphere [6]. It is also expected that these gases to be more concentrated in indoor than outdoor. Exhalation of radon and thoron are mainly associated with the presence of ²²⁶Ra and ²³²Th. The Natural radioactivity in soil sample or building material are usually determined from ²²⁶Ra, ²³²Th and ⁴⁰K contaminant in soil or materials. The average annual indoor effective dose rate of worldwide due to gamma rays from building materials is estimated to be about 0.4 mSvy⁻¹[7]. Studies on building materials such as granite and phosphogypsum enhances indoor absorbed dose rate up to five times than the dose criterion [8], whereas building materials collected from Yan'an, China may be used safely as construction materials [9]. Thus, the assessment of the level of natural radioactivity in building materials is thus become important to evaluate any possible risk to human health and develop any precautionary measures in using different types of building materials.

The study area namely Thoubal district is in the southern side of the state, Manipur and it is in the North Eastern part of India. Thoubal district is having an area of 514 sqkm with total population of 4,20,517 (2011 census).

MATERIALS AND METHODS

Measurement of radiation dose rate (survey meter):

Indoor and outdoor terrestrial gamma radiation of different types of houses in Thoubal district were *Eur. Chem. Bull.* 2022 *11(Regular Issue 10), 446 - 451*

measured by using NaI(Tl) scintillator based micro-Roentgen Survey Meter (SM), manufactured by Nucleonix Systems Pvt. Ltd., Hyderabad, India, having a sensitivity of $1\mu R / hr$. It is measured by maintaining the SM at a height of about 1m distance from the ground surface and wall. Repeated measurements were taken, not less than ten times for each spot. The average value for all these ten measurements is assumed to be the natural gamma radiation dose rate for that particular area [10]. Two hundred twenty-two (222) houses from this district were randomly selected for the evaluation of natural gamma radiation dose rates.

Radiological Analysis of building materials

A total of 10 (ten) samples of building materials from study area were collected and crushed into small sizes. The samples were allowed to dry, in a hot air oven at 110°C for 24 hours. It is then, ground into fine powder, homogenized and sieved through a mesh size of 0.45mm [11], approximately 250 g of this homogeneous fine mesh of each sample material was then packed inside a plastic container with predefined geometry (dimension of cylindrical plastic container: 3" diameter X 5" height), weighted, and properly sealed to restrict the escape of radon or thoron gas from the packed. The processed samples were then stored, carefully for a period of about one month to achieve the equilibrium condition of ²²⁶Ra and ²³²Th along with their respective daughter nuclides [10].

A 3"X3" NaI(Tl) scintillation detector based Gamma spectrometer was used with adequate shielding (about 10 cm lead). The efficiency calibration of the gamma spectrometer was made using different energy peaks covering the range up to \approx 2000 keV. Measurements were performed using calibrated standard source samples, having known activity of gamma ray emitters radionuclides namely ¹³³Ba (356.1 keV), ¹³⁷Cs (661.6 keV), 60Co (1173 KeV and 1332 KeV) and ²²⁶Ra (1764.5 keV) [12]. All prepared samples were subjected to gamma spectral analysis with a counting time of about 36,000 seconds [13]. The concentration of ²²⁶Ra was determined from the average activity concentration obtained from the prominent gamma lines of ²¹⁴Bi (1.76MeV) and ²¹⁴Pb (0.35MeV) and that of ²³²Th was evaluated from the average concentration obtained from the gamma lines of ²¹²Bi (0.73MeV), ²²⁸Ac (0.91MeV) and ²⁰⁸Tl (2.61MeV) respectively; however, ⁴⁰K was evaluated from its own gamma photo-peak (1.46MeV)[11, 14-15].

The activity of ²²⁶Ra, ²³²Th series, and ⁴⁰K were calculated using the following equation [16];

$$A(Bqkg^{-1}) = \frac{N}{\epsilon\beta M}$$
(1)

Where, N = the net gamma counting rate (counts per second), ε = the detector efficiency of the specific gamma-ray, β = the absolute transition probability of gamma decay and M = the mass of the sample (kg).

The concentration and distribution of ²²⁶Ra, ²³²Th and ⁴⁰K is not uniform in the environment as well as in building materials. Radium Equivalent (Ra_{eq}) is a single quantity, most commonly used to represent natural radioactivity associated to those materials containing ²²⁶Ra, ²³²Th and ⁴⁰K. The activity Estimation was made on the assumption that 370.0 BqKg⁻¹ of ²²⁶Ra, 259.0 BqKg⁻¹ of ²³²Th and 4810.0 BqKg⁻¹ of ⁴⁰K produce similar gamma dose rate as mentioned by the following equation [17]

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$
(2)

Where, A_{Ra} , A_{Th} and A_K are the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K in Bqkg⁻¹, respectively.

Activity concentration index, or gamma index 'I', is defined, to examine the applicability of using building materials in construction. It is defined by the following expression [18] as

 $I = A_{Ra} / 300 + A_{Th} / 200 + A_K / 3000 \le 1$ (3)

This is a simple criterion used for the applicability of a building material.

The absorbed Dose Rate (D) due to natural gamma radiation in air for a standard room dimensions and for common building materials with naturally occurring radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K was calculated using the guidelines given in EC report 112[18] as

The annual effective dose rate (D_{eff}) is calculated by applying the dose conversion factor of 0.7 SvGy⁻¹ from the absorbed dose in air received by an adult and a value of 0.8 as an indoor occupancy factor [7]. The annual effective dose rate (D_{eff}) due to gamma radiation from building materials was evaluated as

$$D_{eff} (mSv/y) = D (nGyh^{-1}) X 8760h X 0.8 X 0.7SvGy^{-1} X 10^{-6}$$
(5)

RESULTS AND DISCUSSION:

The natural background gamma radiation levels of 222 different houses

of this Thoubal district were measured using SM. These houses may be classified into 5 different types, based on building materials utilized, which are derived from soils and rocks available in and around the valley region of Manipur state. The classified houses are i) Reinforced Cement Concrete (RCC) House, ii) Assam Type (AT) Katcha House, iii) AT Mud House, iv) AT Brick House and v) Adobe Laid (AL)Earthen House (an indigenous - traditional house of Manipur) so called as Meitei Yumjao and are illustrated in Figure 1. The variation of D_{eff} due to natural gamma radiation level in outdoor as well as indoor of these various types of houses are shown in Figure 2 and detail measured data with error are given in Table 1. It shows that average annual effective dose in indoor of RCC house is 1.2 mSv, followed by AL Earthen houses with a value of 1.1 mSv, AT Brick with a value of 1.0 mSv and least with AT Katcha and AT Mud houses, with values less than unity. Indoor Deff of RCC house is slightly higher than the world average exposure of 0.48 mSvy⁻¹ [19-20]. Whereas, average D_{eff} of outdoor of these houses are almost similar with average value of $0.73 \pm 0.02 \text{ mSvy}^{-1}$.



i) Typical picture of RCC House



ii) Typical picture of Katcha house



iii) Typical picture of AT mud house

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iv) Typical picture of AT brick house



v) Typical picture of AL earthen house (Meitei Yumjao)

The radioactivity concentration values of ²²⁶Ra, ²³²Th and ⁴⁰K measured in the building materials are shown in Table 2. All the Portland cements (3 numbers) and sand-3 (1 number) are imported from neighbouring states. The highest values for specific activity of ²²⁶Ra, ²³²Th and ⁴⁰K are 70.2 Bqkg⁻¹(sand-2 & Soil [21]), 211.8 Bqkg⁻¹(Sand-3) and 2298.6 Bqkg⁻¹ (Cement-3) respectively. The

average specific activity of 226 Ra, 232 Th and 40 K are 43.0 Bqkg^-1 (range: 24.1 - 70.2), 108.6 Bqkg^-1 (range: 21.3 – 211.8) and 1487.2 Bqkg⁻¹(range: 998.7 - 2298.6). The average specific activity of ²²⁶Ra is slightly higher than world average value 35 Bqkg⁻¹ whereas average value of ²³²Th and ⁴⁰K are moderate higher than corresponding world-wide average values of 30 Bqkg⁻¹and 400 Bqkg⁻¹ respectively [7]. Radium equivalent (Raeqv), Gamma index (I), Adsorbed dose rate (D) and Annual effective dose rate (D_{eff}) are given in Table 2. Ra_{eqv} of the building material samples ranged from 216.3 Bqkg⁻¹ (Cement-1) to 465.9 Bqkg⁻ ¹(Sand-3). Ra_{eqv} of two samples namely Sand-3 (465.9 Bqkg⁻¹), and Brick-2 (433.9 Bqkg⁻¹) are found to be higher than the criterion limit of 370 Bqkg⁻¹ [22]. However, the average value of Ra_{eqv} obtained from samples of building materials is 312.8 Bqkg⁻¹, which is less than the critical value (370 Bqkg⁻¹) and as such, does not pose a radiological hazard when used for construction of buildings. Figure 3 shows the Raeqv with different kinds of building materials of Thoubal district, Manipur.



Figure 2: Effective Dose Rate of Outdoor and Indoor for different types of Houses



Figure 3: Absorbed Dose Rate for various building materials *Eur. Chem. Bull.* **2022** *11(Regular Issue 10), 446 - 451*

Gamma index dealing with the assessment of excess gamma radiation arising from building materials ranged from 0.8 (Cement-1) to 1.7 (Sand-3) with an average value of 1.2, which is marginally higher than the recommended value < 1. [18].

The absorbed Dose Rate due to gamma radiations in air for a standard room dimensions and for common building materials with naturally occurring radio nuclides ²²⁶Ra, ²³²Th and ⁴⁰K ranged from 104.4 nGyh⁻¹ (Cement-1) to 212.1 nGyh⁻¹ (Sand-3) with average value of 147.5 nGyh⁻¹, which is higher than world average (populated weighted) indoor absorbed gamma dose rate of 84 nGyh⁻¹ [7].

The annual effective dose rate conceived from building materials were found as 0.7 mSvy^{-1} from Portland cement, 1.0 mSvy^{-1} each from sand and brick. Moreover, D_{eff} due to gamma radiation from building materials ranged from 0.6 mSvy^{-1}

(Cement-1) to 1.3 mSvy^{-1} (Sand-3) with average value of 0.9 mSvy^{-1} .

CONCLUSIONS

A study of natural radioactivity of different types of houses available in Thoubal district of Manipur was conducted. Indoor effective dose rate of RCC houses (1.2 mSvy⁻¹) is observed slightly higher than the Adobe laid Earthen houses with a value of 1.1 mSvy⁻¹ and Assam type (AT) Brick of 1.0 mSvv⁻¹, and least for AT Katcha and AT Mud houses. The activity concentration of common building materials in this region were found as 43.0 Bqkg⁻¹ (range: 24.1 – 70.2) for ²²⁶Ra, 108.6 Bqkg⁻¹ (range: 21.3 – 211.8) for ²³²Th and 1487.2 Bqkg⁻ 1 (range: 998.7 – 2298.6) for 40 K. However, the average value of Ra_{eqy} obtained from samples of building materials of Thoubal district (312.8 Bqkg-¹) is observed less than the critical value (370 Bqkg⁻¹) and as such, does not pose any radiological hazard when used for construction of buildings.

Table 1: Annual Effective Dose of Different types of house in Thoubal District of Manipur.

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Types of House (number)	Indoor (mSvy ⁻¹)			Outdoor (mSvy ⁻¹)						
	Minimum	Maximum	Average	Minimum	Maximum	Average				
RCC House (73)	1.02 ± 0.07	1.35±0.14	1.23±0.10	0.60 ± 0.07	0.89±0.09	0.76 ± 0.08				
Assam Typed Brick House (35)	0.78±0.06	1.24 ± 0.11	1.03±0.09	0.65±0.10	0.91±0.13	0.74 ± 0.08				
Assam Typed Mud House (96)	0.64±0.07	0.99±0.11	0.82±0.09	0.59±0.07	0.94±0.09	0.72 ± 0.08				
Assam Typed Katcha House (6)	0.66±0.09	0.79±0.09	0.70 ± 0.08	0.65±0.06	0.82 ± 0.08	0.70 ± 0.07				
Adobe Laid Earthen House (12)	0.95±0.08	1.19±0.11	1.09 ± 0.10	0.67±0.06	0.95 ± 0.07	0.75 ± 0.08				

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Samula	Activi	Ra _{eqv}	Ι	D	D _{eff}							
Sample	²²⁶ Ra	²³² Th	40 K	$(Bqkg^{-1})$		(nGyh ⁻¹)	(mSvy ⁻¹)					
	Cement Sample											
Cement - 1	29.1±2.7	61.2 ± 5.6	1294.2±81.3	216.3	0.8	104.4	0.6					
Cement - 2	51.8±6.3	23.7±1.7	1749.4±134.5	220.4	0.9	111.2	0.7					
Cement - 3	33.5±5.8	21.3±1.7	2298.6 ± 154.6	241.0	1.0	124.2	0.8					
Average value	38.1±12.0	35.4±22.4	1780.7±502.9	225.9	0.9	113.3	0.7					
	Sand Sample											
Sand - 1	34.1±2.7	56.9±5.8	1764.8±89.6	251.4	1.0	123.7	0.8					
Sand - 2	70.2±11.5	122.6±7.4	1521.6±84.3	362.7	1.4	169.9	1.0					
Sand - 3	51.4±8.1	211.8±14.3	1449.4±69.5	465.9	1.7	212.1	1.3					
Average value	51.9±18.1	130.4±77.8	1578.6±165.3	360.0	1.4	168.6	1.0					
	Brick Sample											
Brick – 1	29.7±3.6	177.1±9.6	1084.9±76.1	366.5	1.4	165.9	1.0					
Brick – 2	24.1±2.7	196.0±9.8	1682.4±99.6	433.9	1.6	199.7	1.2					
Brick – 3	35.8±7.6	139.1±10.4	998.7±54.9	311.6	1.2	142.2	0.9					
Average value	29.9±5.9	170.7±29.0	1255.3±372.4	370.7	1.4	169.3	1.0					
	Soil Sample											
Average value[21]	69.8±32.3	76.6±48.3	1028.0±551.0	258.5	0.9 *	121.4*	0.7*					

*Recalculated using equations (3), (4) & (5)

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