Journal of Environmental Protection, 2018, 9, 1034-1048
http://www.scirp.org/journal/jep
ISSN Online: 2152-2219
ISSN Print: 2152-2197

Natural Radioactivity Measurement and Assessment of Radiological Hazards in Some Building Materials Used in Bangladesh

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How to cite this paper: Khatun, M.A., Ferdous, J. and Haque, M.M. (2018) Natural Radioactivity Measurement and Assessment of Radiological Hazards in Some Building Materials Used in Bangladesh. *Journal of Environmental Protection*, **9**, 1034-1048.
https://doi.org/10.4236/jep.2018.910064

Received: June 7, 2018
Accepted: September 4, 2018
Published: September 7, 2018

Abstract

The radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in 24 samples of natural and manufactured building materials commonly used in Bangladesh were measured using HPGe gamma ray spectrometer. The results in the present study were compared with the world average and also with the reported data available in literature. The radium equivalent activity, the absorbed dose rate, annual effective dose, external and internal hazard indices, gamma index, alpha index, annual gonadal dose equivalent and excess lifetime cancer risk were also evaluated to assess the potential radiation hazards associated with these building materials. All samples under investigation were found to be within the recommended safety limit and do not pose any significant radiation hazards. This study can be used as a reference for more extensive studies of the same subject in future.

Keywords

Natural Radioactivity, Building Materials, HPGe Detector, Radiation Hazards

1. Introduction

Measurement of natural radioactivity concentrations in building materials has fundamental importance in evaluating significant gamma dose indoors, due to their natural radionuclide contents. All building materials are mostly composed of rock and soil containing natural radionuclides such as ^{238}U and ^{232}Th decay series and ^{40}K. These natural radionuclides and their decay products, also called terrestrial background radiation (such as ^{214}Pb, ^{214}Bi, ^{212}Pb, ^{208}Tl, ^{228}Ac, etc.), may cause both external exposure due to their direct gamma radiation and internal exposure from radon gas. Various hazard parameters, such as radium equivalent activity (Ra_{eq}), the absorbed dose rate (D), annual effective dose (D_{eff}), external (H_{ex}) and internal (H_{in}) hazard indices, gamma index (I_γ), alpha index (I_α), annual gonadal dose equivalent (AGDE) and excess lifetime cancer risk (ELCR) play a significant role to assess the potential radiation hazards posed by these building materials. In the ^{238}U series, the decay chain segment starting from ^{226}Ra is radiologically the most important and, therefore, reference in the present study has been made to ^{226}Ra instead of ^{238}U. The world-wide average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the earth crust are about 40 Bq·kg^{-1}, 40 Bq·kg^{-1} and 400 Bq·kg^{-1}, respectively [1].

There has been increased trend of public worldwide in using ceramic tile, stone, marble, granite, etc., due to their polished surface, decorative and different attractive colors, as building materials. The ceramic tiles are generally made of a mixture of different raw materials including clays, quartz materials and feldspar that has been pressed into shape and fired at high temperature. The marble, on the other hand, is a metamorphic rock composed of recrystallized carbonate minerals, most commonly calcite or dolomite. It is extracted from the mountains and after mining it is transported to marble factories in various cities. Granite is the best-known igneous rock. It is composed mainly of quartz and feldspar with minor amounts of mica, amphiboles, and other minerals. A common opacifying constituent of glazes, applied to these materials, is zircon that may cause natural radioactivity concentration significantly higher than the average values for building materials [2] [3] [4].

The worldwide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year [5]. Radiation exposure of the population can be increased appreciably by the use of building materials containing this normal levels of natural radioactivity. It has been demonstrated in various studies that, if building materials with high natural radioactivity concentration are employed, dose rates indoors will be elevated accordingly [2]. Knowledge of basic radiological parameters, such as radioactive contents and their activity concentrations in building materials is, therefore, very much important in the assessment of possible radiation exposure of the population, as most people spending approximately 80% of their lifetimes surrounded by building materials at home and/or at the office [6] [7] [8].

The specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the building raw materials and products mainly depend on geological and geographical conditions as well as geochemical characteristics of those materials [1] [9]. To date, a great attention has been paid to determining radionuclide concentrations in building materials in many countries [3] [9]-[19]. However, for Bangladesh, there is only a few experimental data [7] [20] [21] [22] available in literature regarding the radioactivity of building materials. Mollah *et al.* [20] found a somewhat higher level of activity in building materials than in other countries. Roy *et al.* [21] later carried out an extensive study on the radioactivity in various types of brick samples fabricated and used in Dhaka City and its suburbs. The activity levels in brick samples were found to be consistent with some of that reported in literature. Recently, Asaduzzaman *et al.* [7] and Roy *et al.* [22] investigated some building materials (brick, sand and cement). The aim of the present research is not only the determination of natural radioactivity in some building materials but also the evaluation of the radiological hazard parameters such as Ra_{eq}, D, D_{eff}, H_{ex}, H_{in}, I_γ, I_α, AGDE and ELCR. The present results of radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and their associated radiological hazards parameters mentioned above were compared with the available experimental data.

The rest of this paper has been organized as follows. Section 2 outlines the experimental procedure. In Section 3, we have presented and compared our results systematically with available measurements. Section 4 contains the conclusion on the present findings.

2. Experimental Procedure

2.1. Sample Collection and Preparation

A total of 24 samples of 5 different kinds of building materials used for dwelling in Bangladesh were collected from the dealers. Sample preparation and all radioactivity measurements were performed in the Health Physics Division, Atomic Energy Center (AEC), Dhaka, Bangladesh. The building materials investigated are stone 5 samples, sand 5 samples, cement 5 samples, ceramic tiles 7 samples and marble 2 samples. The samples each about 1 kg in weight were dried in an oven at about 110˚C to ensure that moisture is completely removed. Each of the dried samples (except cement) was grounded to fine powder in an agate motor separately. The powdered samples were then sieved using a fine aperture mesh screen (mesh size 2 μm) in order to remove extraneous items like plant material, roots, pebbles etc. and to obtain a fine grained sample that would present a uniform matrix to the detector. Finally, the grounded samples, approximately 250 - 550 gm of each, were transferred to cylindrical plastic-container (6.5 cm diameter × 7.5 cm height). The containers were then labeled properly and sealed tightly, rapped with thick vinyl tapes around their screw necks. The samples were stored for at least 4 weeks before counting in order to attain secular equilibrium.

2.2. Gamma Spectroscopic Measurements

To qualitatively identify the contents of radionuclides in studied building materials and to quantitatively determine their activities, all prepared samples were subjected to gamma spectral analysis with a counting time of 5000 s. Gamma spectroscopic measurements were performed by means of a coaxial ORTEC HPGe detector with a relative efficiency of 28.2% and an energy resolution of 1.67 keV FWHM at the 1332.5 keV peak of ^{60}Co. The detector was employed with adequate lead shielding to reduce the background radiation from various natural radiation sources and to isolate from other radiation sources used in nearby surroundings.

For energy calibration and relative efficiency calibration of the gamma spectrometer some monoenergetic gamma sources ^{137}Cs, ^{60}Co and ^{40}K were chosen due to a wide range of gamma-ray energies emitted over the entire energy range of interest. The content of ^{226}Ra was measured using the characteristic γ lines of its decay products, including those of ^{214}Pb (295 keV), ^{214}Pb (352 keV), ^{214}Bi (609 keV) and ^{214}Bi (1120 keV). Similarly, the gamma-ray lines of ^{212}Pb (239 keV), ^{208}Tl (583 keV), ^{228}Ac (911 keV) and ^{228}Ac (969 keV) were used for ^{232}Th. The activity of ^{40}K was determined from its intensive gamma-line at 1461 keV. No ^{137}Cs line was obtained at 661.7 keV. The activity concentration of individual radionuclides was calculated from the following analytical expression [23]:

$$A\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right)=\frac{N}{\varepsilon_\gamma\times\rho_\gamma\times T_s\times M} \tag{1}$$

where A is the specific activity in Bq·kg^{-1} of each radionuclide in the sample, N is the net number of counts in the resulting photo-peak, ε_γ is the detector efficiency of the specific gamma-ray, ρ_γ is the intensity at the corresponding gamma-ray energy, T_s is the sample counting time in seconds and M is the mass of the sample in kg. Error associated with every calculation was measured by standard deviation equation.

The background spectrum was used to determine the minimum detectable activity concentration (MDAC) of 0.35 Bq·kg^{-1} for ^{226}Ra, 0.64 Bq·kg^{-1} for ^{232}Th and 2.2 Bq·kg^{-1} for ^{40}K at the 95% confidence level. The equal counting time for both background and sample measurement was chosen to minimize the uncertainty in the net counts. The combined uncertainty of the activity concentration was estimated using [7]:

$$\Delta A=A\sqrt{\left(\frac{\Delta N}{N}\right)^2+\left(\frac{\Delta\varepsilon_\gamma}{\varepsilon_\gamma}\right)^2+\left(\frac{\Delta\rho_\gamma}{\rho_\gamma}\right)^2+\left(\frac{\Delta M_s}{M_s}\right)^2+\left(\frac{\Delta T_s}{T_s}\right)^2} \tag{2}$$

where ΔA is the uncertainty of the sample measurement and ΔN, $\Delta\varepsilon_\gamma$, $\Delta\rho_\gamma$, ΔM_s and ΔT_s are the uncertainties of the count rate, efficiency, gamma-ray emission probability, sample weight and counting time, respectively.

2.3. Estimation of Radiation Hazards

To assess the radiological hazards originating from building materials, several hazard indices have been suggested by a number of investigators [2] [12]. These measures include the radium equivalent activity, the absorbed gamma dose rate in the indoor environment and the corresponding annual effective dose, the external and internal hazard indices, the alpha index (internal index), the gamma activity concentration (gamma index), annual gonadal dose equivalent, excess lifetime cancer risk, etc. In the present study, the aforementioned hazard indicators were estimated to evaluate the potential radiation risks arising from the use of the studied building materials.

Radium equivalent activity was calculated through the relation given by Beretka and Mathew [24]

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Kg·Bq^{-1}, respectively. Equation (3) is based on the estimation that 370 Kg·Bq^{-1} of ^{226}Ra, 259 Kg·Bq^{-1} of ^{232}Th and 4810 Kg·Bq^{-1} of ^{40}K each produce an identical γ-ray dose rate.

If a radionuclide activity is known then its exposure dose rate in air at 1 m above the ground can be found using its conversion factor. The conversion factors of ^{226}Ra, ^{232}Th and ^{40}K are 0.427, 0.662 and 0.043 nGy·h^{-1} per Bq·kg^{-1}, respectively [25]. The contribution of terrestrial gamma radiation to absorbed doses in air was thus calculated using the following formula:

$$D\left(\text{nGy} \cdot \text{h}^{-1}\right) = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_K \quad (4)$$

The absorbed dose rate in air at 1 m above the ground surface does not directly provide the radiological risk to which an individual is exposed. The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which can be estimated by taking into account the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. In the present study, a dose conversion coefficient of 0.7 Sv·Gy^{-1} and an outdoor occupancy factor of 0.2 were used as recommended by UNSCEAR [25]. The annual effective dose equivalent was calculated from following equation:

$$D_{eff}\left(\mu\text{Sv} \cdot \text{y}^{-1}\right) = D\left(\text{nGy} \cdot \text{h}^{-1}\right) \times 8760\left(\text{h} \cdot \text{y}^{-1}\right) \times 0.2 \times 0.7\left(\text{Sv} \cdot \text{Gy}^{-1}\right) \times 10^{-3}. \quad (5)$$

The external hazard index H_{ex} was calculated using the model proposed by Krieger [26], assuming thick walls without windows and doors, as

$$H_{ex} = \frac{A_{Ra}}{370\ \text{Bq} \cdot \text{kg}^{-1}} + \frac{A_{Th}}{259\ \text{Bq} \cdot \text{kg}^{-1}} + \frac{A_K}{4810\ \text{Bq} \cdot \text{kg}^{-1}}. \quad (6)$$

The H_{ex} index must be less than unity so that the annual effective dose due to radioactivity in the material will be ≤1.5 mSv·y^{-1}. In addition to H_{ex} index, inhaled radon and its short-lived progeny also represent a risk to the respiratory organs. Internal exposure to radon and its progeny can be quantified using the index H_{in}, which was estimated by the following expression [26]:

$$H_{in} = \frac{A_{Ra}}{185\ \text{Bq} \cdot \text{kg}^{-1}} + \frac{A_{Th}}{259\ \text{Bq} \cdot \text{kg}^{-1}} + \frac{A_K}{4810\ \text{Bq} \cdot \text{kg}^{-1}}. \quad (7)$$

For the utilization of a building material to be considered safe, H_{in} must be less than unity.

To limit the excess gamma radiation originating from building materials, an index, known as gamma index, is defined as a screening tool for categorizing materials used in construction. It is assumed that activity concentrations of 300 Bq·kg^{-1} of ^{226}Ra, 200 Bq·kg^{-1} of ^{232}Th and 3000 Bq·kg^{-1} of ^{40}K each produce the same gamma dose rate. Therefore, for a typical building material, the gamma index can be calculated using the following equation, as recommended by the European Commission [27]:

$$I_\gamma = \frac{A_{\mathrm{Ra}}}{300\ \mathrm{Bq\cdot kg^{-1}}} + \frac{A_{\mathrm{Th}}}{200\ \mathrm{Bq\cdot kg^{-1}}} + \frac{A_{\mathrm{K}}}{3000\ \mathrm{Bq\cdot kg^{-1}}}. \tag{8}$$

For a structural material, the exemption dose criterion (annual effective dose) of 0.3 mSv·y^{-1} corresponds to a gamma index of $I_\gamma \le 0.5$, whereas the upper dose criterion of 1 mSv·y^{-1} is satisfied for $I_\gamma \le 1$ [27].

Excess alpha radiation caused by the inhalation of radon liberated from building materials can be estimated using the alpha index (I_α) given in [2] [12] [14] [28] as:

$$I_\alpha = \frac{A_{\mathrm{Ra}}}{200\ \mathrm{Bq\cdot kg^{-1}}}. \tag{9}$$

Radon exhalation from a given construction material may lead to indoor radon concentrations that exceed the recommended action level of 200 Bq·m^{-3} if the activity concentration of ^{226}Ra in the material exceeds a value of 200 Bq·kg^{-1} [2] [28]. Thus, the safe limit is defined by an alpha index of less than or equal to unity.

The annual gonadal dose equivalent and excess life-time cancer risk due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K were, respectively, calculated using the following formulae:

$$AGDE\left(\mu\mathrm{Sv\cdot y^{-1}}\right) = 3.09A_{\mathrm{Ra}} + 4.18A_{\mathrm{Th}} + 0.314A_{\mathrm{K}}. \tag{10}$$

and

$$ELCR = D_{\mathrm{eff}} \times DL \times RF. \tag{11}$$

Here, DL (= 70 years) is the duration of life and RF is a risk factor in Sv^{-1} *i.e.* fatal cancer risk per Sievert. For stochastic effects, ICRP 60 [29] uses RF = 0.05 for the public.

3. Results and Discussion

3.1. Activity Concentrations

The measured dry weight activity concentrations of the main gamma emitting radionuclides of the ^{226}Ra series, ^{232}Th series and ^{40}K in 5 different kinds of building-material samples are reported in **Table 1**. The activities of the radionuclides are given in Bq·kg^{-1} and the ± values are due to the 1σ variation of counting uncertainties. The mean specific activities are compared in **Figure 1**. As can be seen in **Table 1**, the largest values for the specific activity of ^{226}Ra and ^{232}Th and ^{40}K are 80.1 ± 2.6, 59.0 ± 5.3 and 803 ± 17 Bq·kg^{-1}, respectively, while the lowest values of the specific activity of the same radionuclides are 7.9 ± 4.6, 4.6 ± 3.8 and 138 ± 12 Bq·kg^{-1}, respectively. The mean specific radioactivity in the 5 different building materials shown in **Figure 1** varies from 11.7 ± 5.4 Bq·kg^{-1} (for stone) to 38.1 ± 9.9 Bq·kg^{-1} (for tile), 14.3 ± 6.9 Bq·kg^{-1} (for stone) to 30.6 ± 8.1 Bq·kg^{-1} (for tile) and 240 ± 4 Bq·kg^{-1} (for marble) to 418 ± 10 Bq·kg^{-1}

Table 1. Radioactivity concentrations of ^{226}Ra series, ^{232}Th series and ^{40}K (in Bq·kg^{-1} ± 1σ) in 24 studied building materials.

Sample Type	^{226}Ra series				^{232}Th series				^{40}K (1461 keV)
	^{214}Pb (295 keV)	^{214}Pb (352 keV)	^{214}Bi (609 keV)	^{214}Bi (1120 keV)	^{212}Pb (239 keV)	^{208}Tl (583 keV)	^{228}Ac (911 keV)	^{228}Ac (969 keV)	
Stone									
ST1	5.78 ± 1.99	4.75 ± 1.35	13.17 ± 1.72	28.04 ± 3.03	11.00 ± 2.16	4.89 ± 0.74	25.25 ± 2.59	16.34 ± 2.93	803.07 ± 16.63
ST2	9.03 ± 1.37	14.21 ± 2.08	13.39 ± 1.26	0.00	9.43 ± 1.40	4.45 ± 0.21	18.81 ± 1.70	7.90 ± 2.07	257.16 ± 11.37
ST3	6.16 ± 1.25	10.53 ± 1.78	10.82 ± 1.72	13.03 ± 0.51	9.39 ± 0.99	4.53 ± 0.72	17.65 ± 1.63	16.56 ± 0.85	295.14 ± 9.67
ST4	12.34 ± 1.87	19.27 ± 2.70	2.43 ± 0.61	8.29 ± 1.64	15.91 ± 2.04	15.12 ± 0.62	19.61 ± 0.74	11.97 ± 1.34	503.70 ± 6.11
ST5	2.39 ± 0.62	22.24 ± 1.96	4.98 ± 1.11	5.30 ± 1.26	11.90 ± 1.69	1.86 ± 0.36	29.28 ± 2.22	10.48 ± 1.34	230.33 ± 6.45
Range	7.9 ± 4.6 to 15.8 ± 4.9				12.0 ± 5.9 to 17.4 ± 10.7				230 ± 6 to 803 ± 17
Sand									
SA1	14.44 ± 2.87	10.72 ± 1.60	9.73 ± 1.06	25.66 ± 4.17	1.64 ± 1.87	2.41 ± 0.46	10.08 ± 1.41	4.24 ± 1.34	137.80 ± 12.22
SA2	11.35 ± 2.87	11.36 ± 1.60	9.85 ± 1.06	25.48 ± 4.17	13.97 ± 1.87	7.52 ± 0.46	26.80 ± 1.41	9.79 ± 1.34	497.29 ± 12.22
SA3	3.02 ± 0.50	5.73 ± 0.80	7.91 ± 0.61	4.47 ± 0.76	5.84 ± 1.17	1.64 ± 0.18	11.93 ± 0.89	17.16 ± 1.83	153.46 ± 4.92
SA4	24.12 ± 7.48	13.55 ± 3.07	16.50 ± 1.67	12.64 ± 1.14	23.22 ± 4.55	8.57 ± 0.95	31.17 ± 1.63	31.40 ± 3.05	333.29 ± 5.09
SA5	7.85 ± 4.73	14.60 ± 3.19	16.26 ± 4.30	24.39 ± 2.40	28.88 ± 4.79	9.74 ± 0.95	27.55 ± 1.93	22.12 ± 2.68	298.55 ± 4.92
Range	11.6 ± 2.5 to 18.1 ± 5.5				4.6 ± 3.8 to 23.6 ± 10.7				138 ± 12 to 497 ± 12
Cement									
CE1	7.50 ± 1.87	20.10 ± 3.31	33.60 ± 2.63	12.90 ± 1.64	27.45 ± 3.21	1.56 ± 0.62	3.44 ± 0.74	31.35 ± 1.34	271.20 ± 6.11
CE2	15.63 ± 1.87	19.14 ± 1.96	10.37 ± 0.66	42.54 ± 5.18	11.30 ± 1.87	2.25 ± 0.44	22.03 ± 1.41	22.03 ± 1.46	212.61 ± 8.32
CE3	12.88 ± 1.74	14.21 ± 2.03	20.75 ± 1.87	23.84 ± 3.41	15.16 ± 1.23	7.79 ± 0.46	28.15 ± 2.59	31.58 ± 2.80	259.87 ± 8.32
CE4	43.12 ± 3.86	25.08 ± 2.33	33.33 ± 2.12	46.12 ± 3.92	16.39 ± 0.99	11.59 ± 0.51	30.83 ± 2.30	33.18 ± 3.20	426.23 ± 10.01
CE5	26.91 ± 1.62	25.75 ± 2.52	31.72 ± 3.23	25.59 ± 1.39	21.34 ± 2.80	8.48 ± 0.10	25.51 ± 1.41	38.51 ± 2.80	319.94 ± 11.54
Range	15.0 ± 4.4 to 33.8 ± 9.0				14.4 ± 8.6 to 23.5 ± 12.4				213 ± 8 to 426 ± 10
Ceramic Tile									
CT1	29.04 ± 4.49	13.12 ± 3.93	20.57 ± 1.52	9.46 ± 2.15	23.75 ± 3.68	0.94 ± 0.51	0.00	33.48 ± 3.78	500.39 ± 7.47
CT2	25.64 ± 4.49	6.97 ± 0.61	8.99 ± 1.57	12.34 ± 2.27	11.02 ± 1.58	3.65 ± 0.82	12.94 ± 1.70	16.47 ± 2.80	207.43 ± 4.41
CT3	80.90 ± 9.10	77.10 ± 4.24	82.18 ± 2.53	94.04 ± 3.16	66.00 ± 5.66	21.18 ± 1.13	49.12 ± 2.44	58.66 ± 3.66	517.80 ± 5.94
CT4	41.74 ± 7.35	12.87 ± 3.19	18.64 ± 1.77	21.29 ± 2.53	16.65 ± 4.38	4.38 ± 0.82	12.65 ± 1.78	7.84 ± 2.56	223.73 ± 3.90
CT5	26.93 ± 4.49	28.03 ± 3.50	32.22 ± 1.92	37.63 ± 2.53	30.19 ± 5.14	13.72 ± 0.95	39.86 ± 1.78	27.41 ± 3.53	319.08 ± 5.09
CT6	45.01 ± 8.72	52.59 ± 3.80	58.54 ± 2.17	50.65 ± 3.03	60.45 ± 5.25	17.91 ± 1.08	53.03 ± 2.15	63.40 ± 3.29	475.28 ± 5.09
CT7	25.16 ± 9.72	35.65 ± 7.92	67.11 ± 3.74	28.88 ± 2.65	34.83 ± 5.60	0.00	35.25 ± 2.67	36.02 ± 4.14	655.43 ± 8.32
Range	17.3 ± 11.8 to 80.1 ± 2.6				10.4 ± 5.4 to 59.0 ± 5.3				207 ± 4 to 655 ± 8
Marble									
IM1	33.61 ± 2.87	14.31 ± 2.52	16.10 ± 1.67	16.17 ± 2.15	24.44 ± 1.28	3.07 ± 0.38	17.30 ± 1.78	15.59 ± 2.68	236.61 ± 4.41
IM2	14.93 ± 7.35	12.01 ± 3.13	13.55 ± 1.67	25.20 ± 2.27	9.67 ± 4.09	5.32 ± 0.85	15.79 ± 1.70	21.97 ± 2.44	242.98 ± 4.07
Range	13.5 ± 1.5 to 21.3 ± 10.7				15.8 ± 6.2 to 19.1 ± 4.5				237 ± 4 to 243 ± 4

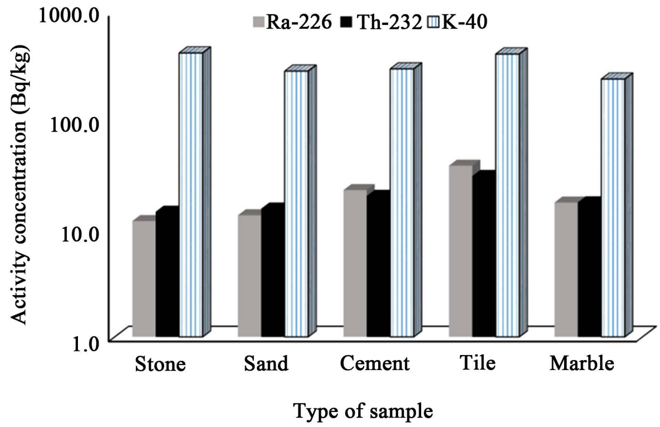

Figure 1. (Colour online) Comparison of mean specific activities of ^{226}Ra, ^{232}Th and ^{40}K measured in 5 different types of studied building materials.

(for stone) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. All these mean values are comparable to the corresponding world average values of 40, 40 and 400 Bq·kg^{-1} [1], respectively for ^{226}Ra, ^{232}Th and ^{40}K. However, the levels of ^{226}Ra in three tile-samples (CT3, CT6 and CT7), ^{232}Th in two tile-samples (CT3 and CT6), and ^{40}K in one stone sample (ST1) and one tile sample (CT7) slightly overestimate the world average.

The comparison of the present results for the ^{226}Ra, ^{232}Th and ^{40}K concentrations with the reported data for selected building materials available in literature are depicted in **Table 2**. The activities for each material and radioisotope show a wide range of values. In general, the mean specific activities of the studied building materials are comparable to those from other countries. However, the ^{226}Ra concentrations in marble samples are much lower in Turkey [3] than the present measurements. For ^{232}Th concentrations, the lower values than the present results are found in Italy [30] for stone samples and in Turkey [3] for marble samples. In case of ^{40}K concentrations for stone samples the present result is higher than those reported by other countries. This variation in activity concentrations may be due to their radioactive mineral content and the geographical origins of the raw materials.

It is worth mentioning that for all studied building materials, the average measured radium activities were found to be greater than the thorium activities. The possible reason for the higher values of uranium is that the Padma river flows, as the Ganges, through Bihar in India, where there is a uranium mine. The Brahmaputra, Surma and Kushiyara rivers flow through Assam in India, where there is a uranium deposit. Thus, it is likely that the traces of uranium and its decay products are carried with the water flow through these rivers, causing the uranium level to be somewhat higher in Bangladesh.

3.2. Hazard Indices

Various hazard indices associated with the radioactivity of the studied building materials and evaluated by using Equations (3)-(11) are presented in **Table 3**.

Table 2. Comparison of activity concentrations (Bq·kg^{-1}) of different building materials from different parts of the world.

Region	Sample Type	^{226}Ra series	^{232}Th series	^{40}K	Reference
Algeria	Stone	16 ± 3	13 ± 2	36 ± 3	[11]
Italy	Stone	11	2.0 ± 2.0	22 ± 3	[30]
Egypt	Stone	27.8 ± 1.4	46.6 ± 2.3	66 ± 3.3	[31]
KSA	Stone	28.6 ± 4.2	49.2 ± 2.5	66 ± 3.6	[17]
India	Stone	46 ± 8	57 ± 12	432 ± 64	[12]
Pakistan	Lime Stone	28.4 ± 8.7	11.3 ± 1.7	63.1 ± 17.3	[32]
Present study	Stone	8 - 16	12 - 18	230 - 803	
Turkey	Sand	38.8 ± 10.0	29.5 ± 11.3	471 ± 101	[14]
Algeria	Sand	12 ± 1	7 ± 1	74 ± 7	[11]
China	Sand	40.7 ± 4.3	21.5 ± 5.6	303 ± 3	[10]
Greece	Sand	18 ± 7	17 ± 10	367 ± 204	[15]
Qatar	Sand	13.2 ± 0.3	3.3 ± 0.1	226 ± 6	[33]
Cuba	Sand	17 ± 4	16 ± 6	208 ± 104	[34]
Present study	Sand	12 - 18	5 - 24	138 - 497	
Turkey	Cement	49.8 ± 5.8	17.3 ± 2.2	246 ± 20	[14]
Greece	Cement	20 ± 5	13 ± 3	247 ± 68	[15]
Algeria	Cement	41 ± 7	27 ± 3	422 ± 3	[11]
Qatar	Cement	23.4 ± 0.6	12.2 ± 0.2	159 ± 4	[33]
Nigeria	Cement	43.8	21.5	72	[19]
Cuba	Cement	23 ± 7	11 ± 3	467 ± 85	[34]
China	Cement	56.5	36.5	173.2	[10]
South Korea	Cement	34.5 ± 1.7	19.4 ± 1.5	241 ± 7	[35]
India	Cement	54 ± 13	65 ± 10	440 ± 91	[12]
Pakistan	Cement	34.2 ± 11.9	29.1 ± 3.6	295.1 ± 66.9	[32]
Present study	Cement	15 - 34	14 - 24	213 - 426	
South Korea	Tile	44 - 82	34 - 96	310 - 1019	[35]
Greece	Ceramics	25 - 174	29 - 47	411 - 786	[13]
Egypt	Ceramics	61 - 118	55 - 98	730 - 1050	[36]
India	Ceramics	28	64	24	[37]
China	Ceramic tile	73	62	480	[4]
Turkey	Ceramic tile	96	53 - 69	290 - 579	[3]
Italy	Porous Tile	53 ± 15	53 ± 12	411 - 996	[16]
Present study	Tile	17 - 80	10 - 59	207 - 655	
Algeria	Marble chip	23 ± 2	18 ± 2	310 ± 3	[11]
Egypt	Marble	205 ± 83	72	870 ± 3.9	[18]
Turkey	Marble	5.4 ± 4.8	4.9 ± 3.8	49.7 ± 19	[3]
Present study	Marble	14 - 21	16 - 19	237 - 243	

Table 3. Calculated various hazard indices associated with the radioactivity of the studied building materials.

Sample	Ra_{eq} (Bq/kg)	D (nGy/h)	D_{eff} (μSv/y)	H_{ex}	H_{in}	I_{γ}	I_{α}	AGDE (μSv/y)	ELCR $\times 10^{-4}$
Stone									
ST1	90.3	45.8	56	0.24	0.27	0.37	0.04	337	11.8
ST2	49.2	23.6	29	0.13	0.17	0.19	0.06	169	5.9
ST3	49.1	23.8	29	0.13	0.16	0.19	0.05	171	6.0
ST4	77.0	37.8	46	0.21	0.25	0.30	0.08	272	9.5
ST5	56.2	26.4	33	0.15	0.19	0.21	0.07	187	6.6
Average	64.4 ± 16.5	31.5 ± 8.9	39 ± 11	0.17 ± 0.04	0.21 ± 0.04	0.25 ± 0.07	0.06 ± 0.01	227 ± 67	8.0 ± 2.3
Range	49 - 90	24 - 46	29 - 56	0.13 - 0.24	0.16 - 0.27	0.19 - 0.37	0.04 - 0.08	169 - 337	6.0 - 12
Sand									
SA1	71.3	35.1	43	0.19	0.23	0.28	0.06	255	8.9
SA2	28.8	13.9	17	0.08	0.11	0.11	0.06	98	3.5
SA3	40.1	18.8	23	0.11	0.14	0.15	0.06	133	4.7
SA4	77.5	36.5	45	0.21	0.26	0.29	0.09	259	9.1
SA5	67.5	31.7	39	0.18	0.22	0.25	0.06	226	7.9
Average	57.0 ± 19.1	27.2 ± 9.1	33 ± 11	0.15 ± 0.05	0.19 ± 0.06	0.22 ± 0.07	0.07 ± 0.01	194 ± 66	6.8 ± 2.3
Range	29 - 78	14 - 37	17 - 45	0.08 - 0.21	0.11 - 0.26	0.11 - 0.29	0.06 - 0.09	98 - 259	3.5 - 9.1
Cement									
CE1	83.3	38.5	47	0.23	0.28	0.31	0.10	208	7.3
CE2	57.8	27.0	33	0.16	0.20	0.21	0.08	190	6.7
CE3	65.5	30.7	38	0.18	0.22	0.24	0.08	217	7.6
CE4	99.6	47.3	58	0.27	0.36	0.37	0.17	335	1.2
CE5	86.3	40.5	50	0.23	0.31	0.32	0.14	285	8.0
Average	78.5 ± 15.0	36.8 ± 7.2	45 ± 9	0.21 ± 0.04	0.27 ± 0.06	0.29 ± 0.06	0.11 ± 0.04	247 ± 55	6.2 ± 2.5
Range	58 - 100	27 - 47	33 - 58	0.16 - 0.27	0.20 - 0.36	0.21 - 0.37	0.08 - 0.17	190 - 335	1.2 - 8.0
Ceramic Tile									
CT1	86.7	42.0	52	0.23	0.29	0.33	0.10	301	10.6
CT2	52.6	24.8	31	0.14	0.19	0.19	0.09	175	6.1
CT3	189.6	88.0	108	0.51	0.73	0.68	0.40	614	21.5
CT4	56.5	26.9	33	0.15	0.22	0.21	0.12	189	6.6
CT5	93.4	43.5	53	0.25	0.33	0.34	0.15	306	10.7
CT6	169.9	77.8	96	0.46	0.60	0.61	0.26	544	19.0
CT7	143.7	68.4	84	0.39	0.50	0.54	0.21	485	17.0
Average	113.2 ± 50.7	53.1 ± 23.2	65 ± 28	0.30 ± 0.14	0.41 ± 0.19	0.48 ± 0.19	0.19 ± 0.10	373 ± 162	13.1 ± 5.7
Range	53 - 190	25 - 88	31 - 108	0.14 - 0.51	0.19 - 0.73	0.19 - 0.68	0.09 - 0.40	175 - 614	6.1 - 21.5
Marble									
IM1	66.9	31.3	38	0.18	0.24	0.25	0.11	220	7.7
IM2	54.8	25.9	32	0.15	0.18	0.21	0.07	184	6.4
Average	60.9 ± 6.1	28.6 ± 2.7	35 ± 3	0.17 ± 0.02	0.21 ± 0.03	0.23 ± 0.02	0.09 ± 0.02	202 ± 18	7.1 ± 0.7
Range	55 - 67	26 - 31	32 - 38	0.15 - 0.18	0.18 - 0.24	0.21 - 0.25	0.07 - 0.11	184 - 220	6.4 - 7.7

Generally, the distribution of ^{226}Ra, ^{232}Th and ^{40}K in environmental samples including construction materials are not uniform. In order to overcome the non-uniformity of the radionuclides, a common index called "radium equivalent activity (Ra_{eq})" is used to obtain the activity and also to assess the radiological hazard caused by the building materials. As shown in **Table 3**, the values of Ra_{eq} varie between 29 and 190 Bq·kg^{-1} with the mean (±SD) values from 57.0 ± 19.1 Bq·kg^{-1} (for sand) to 113.2 ± 50.7 Bq·kg^{-1} (for ceramic tile). It is evident that all the Ra_{eq} values in the present work are lower than the upper recommended value of 370 Bq·kg^{-1} [24]. On comparing with other countries, it is observed that the Ra_{eq} values of this work are lower than that of 436 ± 199 Bq·kg^{-1} for marble and 267 Bq·kg^{-1} for ceramic in Egypt [18] [36]; 183 ± 39 Bq·kg^{-1} for tile in Italy [16]; 121 Bq·kg^{-1} for tile in India [37]; and 112 ± 8.2 Bq·kg^{-1} for cement and 73.0 ± 4.1 Bq·kg^{-1} for marble in Algeria [11]. However, the present results are higher than that of 37.0 ± 4.7 Bq·kg^{-1} for stone and 28.0 ± 7.1 Bq·kg^{-1} for sand in Algeria [11]; and 9.8 ± 3.4 Bq·kg^{-1} for marble in Turkey [3].

The greatest part of the gamma radiation comes from terrestrial radionuclides. There is a direct connection between terrestrial gamma radiation and radionuclide concentrations. The calculated outdoor (terrestrial) gamma dose rates, D in air 1 m above ground range between 24 and 88 nGy·h^{-1}. All of the present D values, except for three tile samples (CT3, CT6, CT7), are lower than the international recommended limit of 57 nGy·h^{-1} [1]. On the other hand, the mean D values range from 27.2 ± 9.1 nGy·h^{-1} (for sand) to 53.1 ± 23.2 nGy·h^{-1} (for tile) and these values are below the world average. The annual effective dose equivalents, D_{eff} calculated from the outdoor terrestrial gamma radiation for the studied 24 building materials are listed in **Table 3**. The values of D_{eff} for the studied samples, except again for three tile samples (CT3, CT6, CT7), are clearly smaller than the world average value of 70 Sv·y^{-1} [1].

Since γ-rays emitted from building materials can easily travel long distances within the surrounding environment, human beings may continuously exposed by gamma radiation and adverse health effects may occurred via extended period of exposure. Thus, the representative gamma-index (I_γ) finds great significance to understand the health hazards from gamma-radiation exposures. Furthermore, external hazard index (H_{ex}) is often used to characterize the building materials to set up a limiting value on the acceptable equivalent dose or to limit the external γ-radiation dose. It is observed in **Table 3** that the mean values of I_γ are below the criterion of 0.5 corresponding to an annual effective dose 0.3 mSv except for the ceramic-tile samples CT3, CT6 and CT7. The mean values of I_γ for these samples (0.68, 0.61 and 0.54 respectively) are below the criterion of unity corresponding to an annual effective dose of 1 mSv. All the present H_{ex} values are lower than the critical value of unity.

Some building materials such as fly-ash and cement can easily be inhaled by people and then the α and β emitters can easily be attached to the living cell of the respiratory organs, causes the cell damage as well as create cancer. For these reasons internal hazard index (H_{in}) and alpha index (I_α) are often used to characterized building materials. As seen in **Table 3**, the present mean values of I_α and H_{in} range from 0.06 ± 0.01 (for stone) to 0.19 ± 0.10 (for ceramic tile) and 0.21 ± 0.03 (for marble) to 0.41 ± 0.19 (for ceramic tile), respectively, and all these values are below the critical value of unity.

The activity bone marrow and the bone surface cells are considered as the organs of interest [25]. Two hazard indices, annual gonadal dose equivalent (AGDE) and associated excess lifetime cancer risk (ELCR), are significant to assess the potential radiation hazard due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K. It is evident from **Table 3** that the mean values of AGDE and ELCR calculated in the present study range from 194 ± 66 to 373 ± 162 $\mu Sv \cdot y^{-1}$ and (6.2 ± 2.5) × 10^{-4} to (13.1 ± 5.7) × 10^{-4}, respectively. On comparing the AGDE values from some other countries, it is observed that value of this work is lower than the calculated values of 550.5 $\mu Sv \cdot y^{-1}$ in Firtna Valley (Rize, Turkey) [38] and 2398 $\mu Sv \cdot y^{-1}$ in Eastern Desert of Egypt [39]; and is comparable to the world-average value of 300 $\mu Sv \cdot y^{-1}$.

4. Conclusion

A total of 24 samples of 5 different kinds natural and manufactured building materials used for dwelling in Bangladesh were analyzed for ^{226}Ra, ^{232}Th and ^{40}K employing gamma spectrometry system equipped with a high-resolution HPGe. The mean concentrations of the above mentioned radionuclides measured in this study were found to be within the typical global range and also compared suitably with the literature values. The measured activity concentrations were also used to estimate several radiological parameters that served to qualify and quantify the radiological hazard associated with the studied building materials. The radium-equivalent activities for the studied building materials were also below the criterion limit of γ-radiation dose of 370 $Bq \cdot kg^{-1}$. The values of internal and external hazard indices for all investigated samples were found to below the unity. The mean annual effective dose equivalents calculated from the outdoor terrestrial gamma radiation for structural building materials were seen to be within the recommended safety limit. The use of these materials in construction of dwellings may, therefore, be considered as safe for inhabitants. As a conclusion, the data reported herein can be used to enlarge the database on natural radioactivity in building materials commonly used in Bangladesh and to support technical aspects in hazard exposure reduction.

Acknowledgements

This study has been carried out within the framework of research project supported by the Faculty of Science, Rajshahi University. The authors wish to thank Prof. Dr. M. Aminul Islam for his kind help.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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