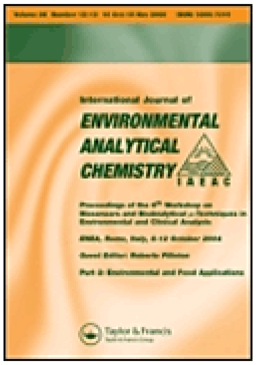

# Assessment of radiometric standard and potential health risks from building materials used in Bangladeshi dwellings

| | |
|---|---|
| Journal: | *International Journal of Environmental Analytical Chemistry* |
| Manuscript ID | GEAC-2021-0221.R2 |
| Manuscript Type: | Original Paper |
| Date Submitted by the Author: | n/a |
| Complete List of Authors: | Rahman, Rubina; University of Liverpool, School of Environmental Science<br>Amatullah, Sajida; Jahangirnagar University Department of Physics, Physics<br>Ferdous, Jannatul; Atomic Energy Centre, Health Physics Division<br>Shiraz, M.M. Mahfuz; Atomic Energy Centre, Health Physics Division<br>Khandaker, Mayeen Uddin; Sunway University, School of Healthcare and Medical Sciences<br>Mahal, Sayeda Ferdous; Atomic Energy Centre, Health Physics Division |
| Keywords: | Radioactivity, Building materials, HPGe Detector, Effective dose, Radiation Hazard Indices |
| | |

# Assessment of radiometric standard and potential health risks from building materials used in Bangladeshi dwellings

S. Amatullah[1], Rubina Rahman[1,3,*], J. Ferdous[2], M.M.M. Siraz[2], Mayeen Uddin Khandaker[4], S.F. Mahal[2]

[1] Department of Physics, Jahangirnagar University, Savar-1342, Dhaka, Bangladesh
[2] Health Physics Division, Atomic Energy Centre, Dhaka-1000, Bangladesh
[3] School of Environmental Science, University of Liverpool, Liverpool, L69 7ZT, UK
[4] Centre for Applied Physics and Radiation Technologies, School of Engineering and Technology, Sunway University, 47500 Bandar Sunway, Selangor Darul Ehsan, Malaysia.

*Corresponding author: Email: rrahman@liverpool.ac.uk; rrahman@juniv.edu

## Abstract

Accurate knowledge of the terrestrial radionuclides concentrations in building components is crucial for radiation exposure assessment to the dwellers. The present investigation determines the natural radioactivity levels in common building materials (the structural and decorative) used in Bangladeshi dwellings via HPGe gamma-ray spectrometry. The measured activity levels of $^{226}$Ra, $^{232}$Th and $^{40}$K in the studied materials ranged between 7.33 ± 3.49 and 157.13 ± 13.03 Bq kg$^{-1}$, 4.08 ± 1.84 and 131.65 ± 6.87 Bq kg$^{-1}$ and 128.38 ± 10.27 and 1234.5 ± 39.77 Bq kg$^{-1}$, respectively. Majority of the studied materials, especially the cement and paint, show the elevated concentrations of terrestrial radionuclides. However, for most of the samples, the total activity in terms of hazardous radium, show lower values compared to the OECD reported limiting index of 370 Bq kg$^{-1}$, except in paint samples. The potential radiological hazards owing to the investigated samples were assessed by calculating a number established screening parameters, and compared with the agreed limits set by international regulatory bodies. The calculated indoor and outdoor absorbed dose rates for most of the materials (apart from sand) overdo the agreeing limiting standards of 84 and 59 nGy h$^{-1}$, as suggested by UNSCEAR (2000). Therefore, continuous radiation level monitoring, especially for paint and cement samples, need to be continued to avoid undesirable exposure from radiation to occupants. Overall, the calculated data may help to set up recommendations for using building materials for dwelling resolutions.

**Keywords:** Radioactivity; Building materials; HPGe detector; Effective dose; Radiation Hazard Indices.

## 1. Introduction

The availability of radioactive materials (NORMs) found in nature is a common feature to be found in our dwelling environment. Among the NORMs, the $^{238}$U, $^{232}$Th and $^{40}$K radionuclides possess the longest half-lives, and found in all soil formations in the trace level while the Earth crust is the main source of these radionuclides. Due to their unstable characteristics, these long-lived radionuclides $^{238}$U and $^{232}$Th undergo a series of decay to achieve the stable form. During the decay process, these radionuclides produce many alpha and beta emitting progeny and subsequently release a range of highly penetrating gamma-rays. Radioactive elements like Radium ($^{226}$Ra), Radon ($^{222}$Rn), Bismuth ($^{214}$Bi), etc. are the decay yields of the Uranium ($^{238}$U) series, and Actinium ($^{228}$Ac), Thoron ($^{220}$Rn), Lead ($^{212}$Pb), Bismuth ($^{212}$Bi), etc. are the decay yields of Thorium ($^{232}$Th) series. Due to their non-negligible presence in all environmental media including air, soil, water, and also in the building materials like brick, sand, cement, wall paint, etc., human being is consciously or unconsciously exposed to ionizing radiation, albeit vary from place-to-place following the native geology and geochemical constituents of the native area [1-6].

Since human being spend almost 80% of their time indoors, the presence of NORMs in our dwelling environment is believed to be the principal source of internal and external radiation exposure to the dwellers. While the internal exposure arises via the inhalation, ingestion and gaseous absorption (exhaled from building elements into indoor air) and metallic radionuclides in the indoor environment, the external exposure occurs via the penetrating gamma rays released from the decay of $^{238}$U, $^{232}$Th and their progeny and $^{40}$K in the building materials [7]. It has been reported that long-term exposure of $^{226}$Ra concentration can create changes in the respiratory system and may cause carcinoma [8-10]. However, it is quite impossible to directly assess the radiation exposure to the dwellers, therefore a hands-on method for this purpose is to evaluate the concentrations of radionuclides in the materials [7]. Hence, the understanding of the naturally available radioactivity in building materials is crucial for an accurate valuation of potential radiation hazards to human health. Therefore, it is vital to measure the concentrations of radionuclides in the materials, used in dwellings construction.

Studies of naturally available radioactivity in building materials and related health risks are conducted in many countries in the world as well as in Bangladesh [11-14]. However, considering the population density and the recent trend of replacement of mud-wood-bamboo based traditional houses by rod-cement-concrete-brick-based structures, the reported data are not enough for a right valuation of radiation exposure to the Bangladeshi dwellers. Moreover, all studies, except the study of [14], are two-three decades back, and such data may not represent the real scenario of the currently used modern building materials. This study also contains some new decorative/ornamental materials that were not used in the earlier studies. It is worth mentioning that different types of building materials may have varied geological origins and mineralogical compositions. Therefore, radiometric analyses of such materials found in diverse regions are crucial for precise evaluation of radiation exposure to the dwellers.

In this connection, this study measures the concentrations of terrestrial radionuclides in a variety of routinely depleted building materials gathered from several dealers in and around the city of Dhaka, Bangladesh. The potential radiological hazards to the dwellers from using such materials in building construction were then assessed by calculating annual effective dose ($E_{eff}$) along with a number of hazard parameters such as the radium equivalent activity ($Ra_{eq}$), external hazard index ($H_{ex}$), internal hazard index ($H_{in}$), alpha index ($I_{\alpha}$), and gamma index ($I_{\gamma}$). The obtained data may find significance in the development of national guidelines for the safe use of materials used in construction of dwellings in accordance of worldwide approvals.

## 2. Materials and methods

### *2.1. Sample collection and preparation*

A total of 31 samples viz. cement, brick, sand, paint, and tiles that are commonly used in Bangladeshi dwellings were collected from local dealers of the capital city, Dhaka, Bangladesh (Table 1). Mass of each sample varied from 0.5 to 1 kg, transferred to individual plastic bags and identified appropriately. The samples were taken to the sample preparation room of Health Physics Division of Atomic Energy Centre Dhaka (AECD), for subsequent processing. They were washed properly (when needed), and dried under the direct sunlight to remove moisture. Samples were further dried using a microwave oven (when needed) to make moisture free and obtain a constant weight. After crushing, powdering, and quartering to a grain size of 1mm (for brick), all samples were then poured and sealed in radon impermeable airtight plastic cans. Then they were stored for 4-6 weeks to achieve secular equilibrium between $^{222}$Rn, $^{220}$Rn and their short-lived daughter yields with the $^{226}$Ra, $^{224}$Ra [15].

### *2.2. Measurement procedures and data analysis*

The activity concentrations of gamma releasing radionuclides within the samples were measured by employing a high-resolution coaxial HPGe gamma-ray spectrometer (EG & G ORTEC) combined with the associated electronics. The effective volume of the detector was 83.47 cm$^3$ and the energy resolution of 1.69 keV at full width half maximum for the 1.33 MeV energy peak of $^{60}$Co, and an efficiency of 19.6 % relative to NaI(Tl) detector. The detector linearity was verified using a $^{152}$Eu gamma-ray-emitting reference source. The energy calibration of the MCA was obtained using standard point sources such as $^{22}$Na, $^{57}$Co, $^{60}$Co, $^{133}$Ba, $^{137}$Cs, etc. The efficiency of the detector for different radionuclides of interest of different energies were determined by mixing $^{152}$Eu of known activity with $Al_2O_3$. The HPGe-detector was enclosed with a lead shield of cylindrical type having a movable cover with fixed bottom to reduce the background contribution from the surrounding environment. Standard sources of solid matrices was made using $^{226}$Ra standard in alike containers to the samples, was used to measure the efficiency of the detector [15]. A MAESTRO 32 multichannel analyzer (MCA) is installed in the computer and coupled to the detector to analyze the acquired gamma-ray spectra and the spectra were evaluated with the computer software program Maestro (EG & G ORTEC) and manually with the use of a spread sheet (Microsoft Excel) to calculate the natural radioactivity.

The radioactivity of $^{226}$Ra and $^{232}$Th were calculated using the representative gamma-lines of their short-lived progeny, as shown in Table 2 [15]. Note that a weighted method [17-18] was adopted for the estimation of $^{226}$Ra and $^{232}$Th radionuclides, and the radioactivity of singly occurring $^{40}$K was directly measured using the net counts under the 1460 keV photo peak. All samples were counted for a satisfactorily long period of 50000[redacted] s and the same counting time were used for the background counts. The net count was then obtained subtracting the background count from the sample count. The activity concentrations of the investigated radionuclides were calculated using Eq. (1) below [19]:

$$A = \frac{N \text{ [redacted]}}{\varepsilon_\gamma \times \rho_\gamma \times T_s \times M_s} \qquad (1)$$

where, A represents the specific activity in Bq kg$^{-1}$, N represents the net number of counts under the characteristic photo-peak, $\varepsilon_\gamma$ is the efficiency of the HPGe coaxial detector at the matching gamma-ray energy, $\rho_\gamma$ represents the branching ratio, $T_s$ is the counting time in seconds and $M_s$ stands for the weight of the sample in kilograms (kg). For the gamma-ray measurement system, the minimum detectable activity concentration (MDAC) was determined using the Eq. (2) as reported in [14]:

$$\mathrm{MDAC} = \frac{K_\alpha \times \sqrt{B}}{\epsilon_\gamma \times \rho_\gamma \times T_s \times M_s} \qquad (2)$$

where $K_\alpha$ is the statistical coverage factor having a value of 1.64 (at the 95% confidence level), B is the number of background counts for the corresponding radionuclide, $\varepsilon_\gamma$, $\rho_\gamma$, $T_s$, and $M_s$ (in kg) have their usual meaning similar to Eq. (1). The MDAC values for the investigated radionuclides in this study were found to be 0.75 Bq kg$^{-1}$ for $^{226}$Ra, 1.08 Bq kg$^{-1}$ for $^{232}$Th, and 0.67 Bq kg$^{-1}$ for $^{40}$K, and the uncertainty of the measured activity concentration was derived using following Eq. (3)

$$\sigma = \sqrt{\left[\frac{N_s}{T_s^2} + \frac{N_b}{T_b^2}\right]} \tag{3}$$

where $N_s$ is the measured counts in time $T_s$ and $N_b$ is the background counts in time $T_b$. The standard deviation (± 2σ), in CPS, was then transformed into activity concentration in Bq kg$^{-1}$.

### 2.3 Radium Equivalent Activity ($Ra_{eq}$) evaluation

The existence of $^{226}$Ra, $^{232}$Th, and $^{40}$K concentrations is evenly distributed in the environment, and eventually they are not so de in the building materials. The non-uniformity of radioactivity in building materials having $^{226}$Ra, $^{232}$Th, and $^{40}$K can be demonstrated using a single index, $Ra_{eq}$ which characterizes both the total activity in terms of hazardous radium, and thus the radiological risk initiated by the building materials. In this study, $Ra_{eq}$ was computed using the following Eq. (4) as stated in [20-21]:

$$Ra_{eq} = 370\left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \tag{4}$$

where, $A_{Ra}$, $A_{Th}$, and $A_K$ (in Bq kg$^{-1}$) are the measured activities of $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively. The above Eq. (4) is founded on the assessment that 370 Bq kg$^{-1}$ of $^{226}$Ra, 259 Bq kg$^{-1}$ of $^{232}$Th, and 4810 Bq kg$^{-1}$ of $^{40}$K each yield an alike γ-ray dose rate [22]. It is recommended that the maximum activity level of 370 Bq kg$^{-1}$ in any materials corresponds to an annual effective dose of 1.5 mSvy$^{-1}$ [23].

### 2.4 Absorbed Dose Rate in Air and Annual Effective Dose evaluation

The external absorbed dose rate, $D_{out}$ owing to the exposure of the emitted gamma-rays from the building material to the public in the outdoor air was determined using the following Eq. (5) [14]:

$$D_{out} = 0.427 \times A_{Ra} + 0.662 \times A_{Th} + 0.0432 \times A_K \tag{5}$$

where, $D_{out}$ is the outdoor absorbed dose rate in (nGy h$^{-1}$) owing to exposure of gamma-rays, and other symbols have their usual meaning. It is expected to have that indoor exposure from gamma rays to man is naturally higher than the outdoor exposure from gamma rays because most of the raw materials used for building construction are extracted from the earth surface media. On the other hand, the duration of occupancy of human being in indoor is much longer than the outdoor, consequently the indoor exposure becomes more noteworthy. Since some materials like brick, sand, cement, paints, tiles etc. that are originated from earth crust are extensively used in construction of inhabitations. It is thus imperative to assess the indoor exposure, and used the Eq. (6) as reported in [6, 14]:

$$D_{in} = 1.4 \times D_{out} \quad (6)$$

The assessed indoor and outdoor exposures can be used to estimate the corresponding annual effective doses $E_{in}$ and $E_{out}$. To do this, a conversion factor of 0.7 Sv $Gy^{-1}$ was used for the conversion of the absorbed dose rate in the air to the effective dose received by an adult [14]. Moreover, since people generally spend about 80% and 20% of their time indoor and outdoor respectively, therefore the values of 0.8 and 0.2 for the indoor and outdoor occupancy factors are used to obtain the representing dose (Sharaf and Hamideen, 2013). Thus, the annual effective doses $E_{in}$ (mSv $y^{-1}$) and $E_{out}$ (mSv $y^{-1}$) were estimated using the following Eqs. (7, 8) [6]:

$$\mathrm{E_{in}}(\mathrm{mSv}\, y^{-1}) = \mathrm{D_{in}}(\mathrm{nGyh}^{-1}) \times (8760\mathrm{h}\, y^{-1} \times 0.7\mathrm{Sv\,Gy}^{-1} \times 0.8) \times 10^{-6}$$

(7)

$$\mathrm{E_{out}}(\mathrm{mSv}\, y^{-1}) = \mathrm{D_{out}}(\mathrm{nGyh}^{-1}) \times (8760\mathrm{h}\, y^{-1} \times 0.7\mathrm{Sv\,Gy}^{-1} \times 0.2) \times 10^{-6} \quad (8)$$

### *2.5 Gamma Index ($I_\gamma$)*

The gamma index is suggested as an inspection parameter for categorizing elements to be used in construction purpose [22]. For this reason, the European Commission recommended formula shown in Eq. (9) is used [24],

$$I_\gamma = \frac{A_{Ra}}{300Bqkg^{-1}} + \frac{A_{Th}}{200Bqkg^{-1}} + \frac{A_K}{3000Bqkg^{-1}} \quad (9)$$

It is considered that activity of 300 Bq $kg^{-1}$ for $^{226}$Ra, 200 Bq $kg^{-1}$ for $^{232}$Th, and 3000 Bq $kg^{-1}$ for $^{40}$K each produces an equivalent gamma dose rate. For the gamma dose of building materials, the European Commission (1999) recommended two criteria: an exemption criterion of 0.3 mSv $y^{-1}$ and the maximum limit of 1 mSv $y^{-1}$. For a structural material like brick, sand, etc., the exemption criterion of 0.3 mSv $y^{-1}$ links to a gamma index of $I\gamma \leq 0.5$, whereas the upper dose criterion of 1 mSv $y^{-1}$ is gratified for $I\gamma \leq 1$ [24]. Furthermore, for the decorative (such as titles) and superficial (paint, board, etc.) building materials, the value of $I\gamma$ should not exceed 2 and 6 based on the annual dose limits of 0.3 mSv $y^{-1}$ and 1 mSv $y^{-1}$, respectively [7].

### *2.6 Alpha Index ($I_\alpha$)*

The exposure from alpha owing to the inhalation of radon gas initiating from building materials can be evaluated through the alpha index (Iα) [25]:

$$I_\alpha = \frac{A_{Ra}}{200 Bqkg^{-1}} \tag{10}$$

where, $A_{Ra}$ is the activity of the $^{226}Ra$ precursor that produces gaseous $^{222}Rn$ (Bq $kg^{-1}$). Radon exhalation from construction materials may be the source of indoor radon concentrations that exceed the recommended set level of 200 Bq $m^{-3}$ if the activity concentration of $^{226}Ra$ within the material outstrips a value of 200 Bq $kg^{-1}$ [25]; hence, the safe boundary is defined to unity for an alpha index.

### *2.7 External Hazard ($H_{ex}$) and Internal Hazard ($H_{in}$) Indices evaluation*

The external and internal hazard indices are useful to line a restrictive value on the acceptable equivalent dose [6] as recommended by the ICRP (1990) [26]. To limit the radiation dose from building materials, the value of $H_{ex}$ need to be less than or equal to unity [22]. Within this study, $H_{ex}$ was determined using the Eq. (11) as conveyed by Beretka and Mathew (1985) [23]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{11}$$

Inhaled radon and its short-lived progenies also exemplify a hazard to the respiratory organs. Internal exposure to radon and its progeny are often counted using the index $H_{in}$, which is assessed using the subsequent Eq. (12) [23, 25]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{12}$$

For the use of building materials to be encountered safely, $H_{in}$ need to be ≤1 [22,27].

## 3. Results and Discussion

The activity concentrations of $^{226}Ra$, $^{232}Th$, and $^{40}K$ in the studied building materials are presented in Table 3. For the investigated samples, it is found that the activity concentration of $^{226}Ra$ ranges from 7.33 ± 3.49 to 157.13 ± 13.03 Bq $kg^{-1}$ with a mean of 53.06 ± 5.05 Bq $kg^{-1}$, $^{232}Th$ ranges from 4.08 ± 1.84 to 131.65 ± 6.87 Bq $kg^{-1}$ with a mean of 43.69 ± 4.37 Bq $kg^{-1}$, and $^{40}K$ ranges from

128.38 ± 10.27 to 1234.5 ± 39.77 Bq kg$^{-1}$ with mean of 590.79 ± 20.43 Bq kg$^{-1}$. No peak of artificial fission fragment $^{137}$Cs (662 keV) was observed within the gamma-ray spectrum. It is going to be concluded that either there is no $^{137}$Cs radionuclide within the investigated samples or the $^{137}$Cs activity is below the detection limit of 1.54 Bq kg$^{-1}$ (for $^{137}$Cs) in the measurement system in the present study.

It is a common scenario that the concentration of $^{40}$K show the highest value in any geological material [15], and this phenomenon is also observed in the present building materials. Among the studied building materials, a relatively higher mean activity concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th are observed in the cement and paint samples (see in Table 3). Note that, in addition to the locally made cement and paint in the country, some available brands (like Jhilik paint, sample P4) are imported from other countries. Cement is manufactured through a closely controlled common materials including limestone, shells, chalk, clay, slate, blast furnace slag, sand, and iron ore. Though most of the cements in the present study are manufactured in Bangladesh, however, some of the above mentioned raw materials are usually imported from other countries. Consequently, the radioactivity in cement varies following the geological origin of the aforementioned ingredients used in their production. Hence, this could be a reason for higher activity concentration in some cement samples (like samples C2 and C4; Table 3) in this study. Moreover, some materials like fly ash is also used as a supplementary cementing material (SCM) which contributes to the properties of the hardened concrete, and such SCM contains relatively high concentration of NORMs. On the other hand, the paints are made from the mixture of earthly components such as pigments, binders, solvents and various additives and are glazed by adding the zircon pigments. It has been established that zircon pigments or other earthly components are characterized by the high level of NORMs, as a result, paints may be found to possess a high level of radioactivity [28]. Furthermore, the relatively high levels of radioactivity in the bricks, sand, and tiles may be due to the local geology since their raw materials are products of the Earth Crust.

A comparison of the average activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K for the analyzed samples with the available literature is presented in Table 4. It shows that the typical activity concentration of $^{226}$Ra (53 Bq kg$^{-1}$) and $^{232}$Th (44.01 Bq kg$^{-1}$) are above the values of Greece, Italy, Turkey, and Pakistan and less than the values of China, Egypt, India and former studies in Bangladesh. The typical activity concentration of $^{40}$K (591 Bq kg$^{-1}$) is above all other included countries but less than the previous study from Bangladesh. However, the overall mean values for $^{40}$K, $^{226}$Ra and $^{232}$Th radionuclides are found to be 591, 53 and 44 Bq kg$^{-1}$ respectively, and these values exceed (except $^{232}$Th) the corresponding world average values of 500, 50, and 50 Bq kg$^{-1}$ for building materials [29].

Table 5 shows the radiological parameters estimated for the investigated samples under this study. The Ra$_{eq}$ ranged from 28 Bq kg$^{-1}$ in (White Sand) to 385 Bq kg$^{-1}$ in (Paint) with an overall average of 161 Bq kg$^{-1}$. Most of the values of Ra$_{eq}$ in the studied samples is lower than the OECD recommended standard limit of 370 Bq kg$^{-1}$, except for a few paint samples. This indicates that the use of paint should be limited or subject to be perpetual monitoring.

The estimated outdoor absorbed dose rate (D$_{out}$) for cement, brick, white sand, red sand, paint, and tiles were found as 101, 73, 47, 58, 114, and 69 nGy h$^{-1}$, respectively, with the mean value of 77 nGy h$^{-1}$. On the other side, the estimated indoor absorbed dose rate (D$_{in}$) for cement, brick, white sand, red sand, paint, and tiles were found to be 121, 88, 56, 70, 137, and 83 nGy h$^{-1}$, respectively, with a mean of 93 nGy h$^{-1}$ (Table 5). However, most of the calculated values of D$_{out}$ (except for the sand samples) are greater than the recommended limit of 59 nGy.h$^{-1}$, as suggested by UNSCEAR (2000) [6]. Furthermore, majority of the samples (except sand and titles) show values of D$_{in}$ to be higher than the criterion limit of 84 nGy h$^{-1}$, as suggested by UNSCEAR (2000) [6].

The outdoor annual effective dose (E$_{out}$) values due to the emitted gamma radiation from the cement, brick, white sand, red sand, paint, and tiles samples were found to be 0.12, 0.09, 0.05, 0.07, 0.14, and 0.08 mSv y$^{-1}$, respectively, with a mean value of 0.10 mSv y$^{-1}$ (Table 5; Fig. 1 and Fig. 2), which is 43% higher than the world average of 0.07 mSv y$^{-1}$ [6]. On the other hand, the indoor annual effective dose (E$_{in}$) values due to the emitted gamma radiation from the cement, brick, white sand, red sand, paint, and tiles samples were found to be 0.72, 0.52, 0.33, 0.41, 0.81, and 0.49 mSv y$^{-1}$, respectively with a mean value of 0.55 mSv y$^{-1}$ (Table 5; Fig. 1 and Fig. 2), which is 34% higher than the world average of 0.41 mSv y$^{-1}$ [6]. This indicates that a prolonged both outdoor and indoor exposures to gamma radiation from these materials may pose non-negligible health hazards. However, these are lower than the European Commission (1999) suggested total value of 1 mSvy$^{-1}$ coming from the sum of outdoor and indoor exposure to the gamma radiation. On the basis of both outdoor and indoor annual effective dose criterions, all materials (except sands) should be used in a controlled manner to reduce the gamma exposure to dwellers.

Moreover, a clear picture on the relative $^{226}$Ra, $^{232}$Th, and $^{40}$K contributions to the annual effective indoor dose ($E_{in}$) and annual effective outdoor dose ($E_{out}$) (see in Fig. 1), and the dose distributions in terms of building materials and individual radionuclides are presented in Figs. 2 (a, b). It is seen that the contribution of indoor annual effective doses is 83% whereas the outdoor annual effective dose contributes to 17% of the rest of the total annual effective dose (Fig. 1). Furthermore, Figure 2 shows a relative annual effective dose distribution due to $^{226}$Ra, $^{232}$Th, and $^{40}$K contributions in investigated building materials with an ascending order of effective dose rate as

Paint > Cement > Brick > Tiles > Sand.

Another concern is that paints are used at the most upper part on both sides of the wall and under side of the roof (as furnishing material), thus it poses more health hazard risk compared to others materials. Hence, more investigations are needed especially on paint and cement samples including associated raw materials of these products to make a precise conclusion on the existing higher radioactivity levels in these materials, available in Bangladesh. Moreover, priority should be given on the building materials those are safe from radioactivity point of view, to maintain a safe living environments for the human beings.

In regard to the hazard indices, the calculated values for $H_{ex}$ and $H_{in}$, $I_\alpha$ and $I_\gamma$ indices for all investigated building materials were below the limit of unity, meaning that the radiation dose is below the maximum dose limit of 1 mSv $y^{-1}$ recommended by ICRP [26, 30]. Note that the use of internal hazard indices such as $H_{in}$ and $I_\alpha$ are often used to characterize building materials. This is because some of the studied materials like cement can be inhaled by the workers, and the α and β emitters can easily be attached to the respiratory organs, and create unexpected exposure.

## 4. Conclusion

The activity concentrations of naturally occurring radionuclides in building materials such as brick, sand, cement, paint, and tiles to be used in Bangladeshi dwellings are measured by HPGe gamma-ray spectrometry. The measured mean activity concentrations of 591, 53 and 44 Bq $kg^{-1}$ for $^{40}$K, $^{226}$Ra and $^{232}$Th found to be higher (except $^{232}$Th) than the UNSCEAR reported world average values of 500, 50, and 50 Bq $kg^{-1}$ respectively. In general, the paint and cement samples show the higher level of radioactivity while the white sand shows the lowest values. Both the outdoor and indoor absorbed dose rates for all samples (except sand) exceed the criterion limits of 59 nGy.$h^{-1}$ and 84 nGy $h^{-1}$, suggested by the UNSCEAR. Similarly, the annual effective dose for all samples show 43% and 34% higher than the world average of 0.07 mSv $y^{-1}$ and 0.41 mSv $y^{-1}$ for outdoor and indoor doses, respectively. It has been found that $^{40}K$ is the largest dose contributor to the total effective dose. However, the overall average values of other radiological indices do not exceed their corresponding upper limits, which are, $Ra_{eq} < 370$ (except for a few paint samples), $H_{ex}$, $H_{in}$, $I_{\alpha}$, and $I_{\gamma}$, < 1, which derived based on an annual effective dose of 1 mSv $y^{-1}$. The estimated dose and other parameters indicate that some of the materials (especially the paint and cement samples) should be placed under perpetual monitoring when they are used as building materials to avoid any unnecessary exposure to radiation. It is expected that, the data of this study can be used as a reference for any future radiological studies of construction materials in Bangladesh.

## Acknowledgement

The authors would like to thank to the Director, Atomic Energy Centre, Dhaka, (AECD), and the Head, Health Physics Division, AECD, Bangladesh to give permission to provide their research facilities to complete this work.

## Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Reference:

1. Kh. Asaduzzaman, M.U. Khandaker, Y.M. Amin, D.A. Bradley, R.H., Mahat, R.M. Nor, J. Environ. Radioact. 135, 120 (2014).
2. Kh. Asaduzzaman, M.U. Khandaker, Y.M. Amin and D.A. Bradley, Indoor Built Environ.25 (3), 541(2016). https://doi.org/10.1177/1420326X14562048.
3. N.M. Hassan, N.A. Mansour, M. Fayez-Hassan, Radiat. Protect. Dosim. 157, 214 (2013). https://doi.org/10.1093/rpd/nct129.
4. M.T. Kolo, S.A. Aziz, M.U. Khandaker, Kh. Asaduzzaman, and Y.M. Amin, 22 (17), 13127 (2015).
5. M.U. Khandaker, N.L.M. Nasir, N.S. Zakirin, H.A. Kassim, Kh. Asaduzzaman, D.A. Bradley, M.Y. Zulkifli, A. Hayyan, Radiat. Phys. Chem. 140, 173 (2017).
6. UNSCEAR 1993 (United Nations Scientific Committee on the Effects of Atomic Radiation) Annex. B. Exposures from natural radiation sources, UNSCEAR (2000).
7. K. Aladeniyi, A.M. Arogunjo, A.J.S.C. Pereira, M. U. Khandaker, D.A. Bradley, Radiat. Phys. Chem. 178, 109021 (2021).

8. Raghu, Y., Ravisankar, R., Chandrasekaran, A., Vijayagopal, P., Venkatraman, B., J. Taibah Univ. Sci.,11 (4), 523 (2017).
9. Kh. Asaduzzaman, M. U. Khandaker, Y. M. Amin, R. Mahat, Ann. Nucl. Energy, 76, 85 (2015a).
10. G. Senthilkumar, Y. Raghu, S. Sivakumar, A. Chandrasekaran, D. Prem Anand, R.J. Ravisankar, Radiat. Res. Appl. Sci., 7 (1), 116 (2014).
11. A.S. Mollah, G.U. Ahmed, S.R. Husain and M.M. Rahman, Health Phys; 50: 849 (1986).
12. I. M. Chowdhury, M.N. Alam and A.K.S. Ahmed, J. Radioanal. Nucl. Chem. 231, 117 (1998).
13. S. Roy, M.S. Alam, M. Begum and B. Alam, Radiat. Prot. Dosim., 114, 527 (2005).
14. Kh. Asaduzzaman, F. Mannan, M.U. Khandaker, M.S. Farook, A. Elkezza, Y.B. Amin, S. Sharma, H.B. Abu Kassim, PLoS One, 10 (10), 1 (2015b).
15. M.M. Hasan, A.H. Chaity, M.A. Haydar, M.I. Ali, M.U. Khandaker, Indoor Built Environ. (2020). https://doi.org/10.1177/1420326X20924835.
16. M.U. Khandaker, N.L.M. Nasir, Kh. Asaduzzaman, M.A. Olatunji, Y.M. Amin, H.A. Kassim, D.A. Bradley, P.J. Jojo, T. Alrefae, Chemosphere 154, 528 (2016).
17. M.U. Khandaker, N.A.B. Heffny, Y.M. Amin, D.A. Bradley, Algal Res. 38, 101386, (2019a).
18. M.U. Khandaker, O.B. Uwatse, K.A. Bin Shamsul Khairi, M.R.I. Faruque, D.A. Bradley, Radiat. Protect. Dosim. 185, 343 (2019b).
19. M.U. Khandaker, N.B. Wahib, Y.M. Amin, D.A. Bradley, D.A., Radiat. Phys. Chem. 88, 1 (2013).
20. H.K. Shuaibu, M.U. Khandaker, T. Alrefae, D.A. Bradley, Mar. Pollut. Bull., 119 (1), 423(2017).
21. M.U. Khandaker, Kh. Asaduzzaman, A.F. Sulaiman, D.A. Bradley, M.O. Isinkaye, Mar. Pollut. Bull. 127, 654 (2018).
22. S. A. Amin, M. Naji, Radiat. Phys. Chem., 86, 37 (2013).
23. J. Beretka, P.J. Mathew, Health Phys., 48 (1), 87 (2014).
24. M. Gupta, A.K. Mahur, R. Varshney, R.G. Sonkawade, K.D. Verma, R. Prasad, Radiat. Meas.50, 160 (2013).
25. M.A. Kobeissi, O. El-Samad, I. Rachidi, Radiat. Prot. Dosim. 153 (3), 342 (2013).
26. ICRP International Commission on Radiological Protection. Publication 60, 21, Ann. ICRP 21(1-3), Pergamon Press, Oxford (1990).
27. J.M. Sharaf, and M.S. Hamideen, Appl. Radiat. Isot., 80, 61 (2013).
28. S. Yasmin, M.U. Khandaker, B.S. Barua, M.N. Mustafa, F.U.Z. Chowdhury, M.A. Rashid, D.A. Bradley, D.A., Indoor Built Environ. 28 (6), 825(2019).
29. UNSCEAR 1993 (United Nations Scientific Committee on the Effects of Atomic Radiation)

Sources and effects of ionizing radiation. New York, NY: United Nations Publication, (1993).

30. N. Krieger, American J. Public Health. 107 (4), 541(2017).
31. M.U. Khandaker, P.J. Jojo, H.A. Kassim, Y. M. Amin, Radiat. Protect. Dosim. 152, 33 (2012).
32. S.U. Rahman, M. Rafique, A. Jabbar, and Matiullah, Radiat. Prot. Dosimetry. 153 (3): 352 (2013).
33. S. Solak, S. Turhan, F.A. Ugur, E. Goren, F. Gezer, Z. Yegingil, et al. Indoor Built Environ 23(4): 594 (2014).
34. L. Xinwei, Radiat. Meas. 40, 94 (2005).
35. N. K. Ahmed, J. Environ. Radioact. 83. 91 (2005).
36. S. Righi, and L. Bruzzi, J. Environ. Radioact. 88, 158 (2006).
37. S. Stoulos, M. Manolopoulou and C. Papastefanou. J. Environ. Radioact. 69, 225 (2003).

**Table 1:** Basic information of the investigated building materials used in Bangladeshi Dwellings. The symbol (W) and (R) refers to white- and red sand.

| Serial no. | Sample ID | Sample name | Manufacturer/Dealer/Supplier | Country of Origin/ Production |
|---|---|---|---|---|
| 1. | C1 | Seven Ring cement | Seven Circle Bangladesh Ltd. | Bangladesh |
| 2. | C2 | Tiger cement | Madina Cement Industries Ltd. | Bangladesh |
| 3. | C3 | Montana cement | S.C.T Co. Ltd. | Thailand |
| 4. | C4 | Elephant Brand cement | Siam Bangla Ltd. | Bangladesh |
| 5. | C5 | M. G. Gourt cement | M.G Grout Ltd. | Bangladesh |
| 6. | C6 | Bashundhara cement | Basundhara Industries Ltd. | Bangladesh |
| 7. | B1 | Brick 1 | Ashulia, Dhaka | Bangladesh |
| 8. | B2 | Brick 2 | Ashulia, Dhaka | Bangladesh |
| 9. | B3 | Brick 3 | Ashulia, Dhaka | Bangladesh |
| 10. | B4 | ASB Brick | Kalampur, Dhamrai, Dhaka | Bangladesh |
| 11. | B5 | ABC Brick | Kalampur, Dhamrai, Dhaka | Bangladesh |
| 12. | B6 | Brick 4 | Gazipur, Dhaka | Bangladesh |
| 13. | S1(W) | White sand 1 | Kaliyakaur. Balughat, Gazipur | Bangladesh |
| 14. | S2(W) | White sand 2 | Kuturiya, Ashulia, Dhaka | Bangladesh |
| 15. | S3(W) | White sand 3 | Dhamrai, Dhaka | Bangladesh |
| 16. | S4(W) | White sand 4 | Gabtoli, Dhaka | Bangladesh |
| 17. | S5(W) | White sand 5 | Gabtoli, Dhaka | Bangladesh |
| 18. | S6(R) | Red sand 1 | Sunamgaung, Sylhet | Bangladesh |
| 19. | S7(R) | Red sand 2 | Sunamgaung, Sylhet | Bangladesh |
| 20. | S8(R) | Red sand 3 | Sunamgaung, Sylhet | Bangladesh |
| 21. | P1 | RAK paint | RAK paint Ltd. | Bangladesh |
| 22. | P2 | Polac plastic paint | Polac paint and Chemical Co. Ltd. | Bangladesh |
| 23. | P3 | Polac synthetic enamel | Polac paint and Chemical Co. Ltd. | Bangladesh |
| 24. | P4 | Berger Jhilik | Berger Paints India Ltd. | India |
| 25. | T1 | Fu-Wang tiles | Fu-wang Ceramic Industries Ltd. | Bangladesh |
| 26. | T2 | Mir titles | Mir Ceramic Ltd. | Bangladesh |
| 27. | T3 | Sun power tiles | Sun Power Ceramic Ltd. | Indonesia |
| 28. | T4 | Akij tiles | Akij Ceramic Industries Ltd. | Bangladesh |
| 29. | T5 | Great wall | Great Wall Ceramic Industries Ltd. | Bangladesh |
| 30. | T6 | ABC tiles | ABC Ceramic India | India |
| 31. | T7 | DSC tiles | Johnson Floor Company Ltd. | China & Bangladesh |

**Table 2:** Decay data of radionuclides of interest used for calculations of radioactivity in building material [31].

| Radionuclides of interest | Detected radionuclides | Half-life | Decay mode (%) | γ-ray energy, $E_\gamma$ (keV) | Branching ratio, $I_\gamma$ (%) | MDA (Bq kg$^{-1}$) | Sources/origin |
|---|---|---|---|---|---|---|---|
| $^{226}$Ra | $^{214}$Pb | 26.80 m | $\beta^-$ (100) | 295.22 | 18.42 | 1.08 | $^{238}$U ($^{226}$Ra) series |
| | | | | 351.93 | 35.6 | 0.61 | |
| | $^{214}$Bi | 19.90 m | α (0.02); $\beta^-$ (99.98) | 609.32 | 45.49 | 0.72 | $^{238}$U ($^{226}$Ra) series |
| | | | | 1120.294 | 14.92 | 0.87 | |
| | | | | 1764.491 | 15.3 | 0.47 | |
| | | | | | **Mean** | **0.75** | |
| $^{232}$Th | $^{228}$Ac | 6.15 h | α+ $\beta^-$ (100) | 911.204 | 25.8 | 0.76 | $^{232}$Th series |
| | | | | 968.971 | 15.8 | 1.21 | |
| | $^{212}$Pb | 10.64 h | $\beta^-$ (100) | 238.632 | 43.6 | 1.91 | $^{232}$Th ($^{228}$Ra) series |
| | $^{208}$Tl | 3.053 m | $\beta^-$ (100) | 583.187 | 85 | 0.44 | $^{232}$Th ($^{228}$Ra) series |
| | | | | | **Mean** | **1.08** | |
| $^{40}$K | $^{40}$K | 1.248E+09 y | EC (10.72); $\beta^-$ (89.28) | 1460.822 | 10.66 | **0.67** | Primordial/terrestrial |

**Table 3:** Activity concentrations (Bq kg$^{-1}$) of radionuclides in the studied building materials commonly used in Bangladeshi dwellings.

| Serial no. | Sample ID | $^{226}$Ra (Bq kg$^{-1}$) | $^{232}$Th (Bq kg$^{-1}$) | $^{40}$K (Bq kg$^{-1}$) |
|---|---|---|---|---|
| 1. | C1 | 65.32 ± 4.46 | 27.81± 3.65 | 623.55 ± 22.07 |
| 2. | C2 | 126.57 ± 8.08 | 110.40 ± 6.44 | 720.47 ± 23.85 |
| 3. | C3 | 66.80 ± 5.98 | 41.75 ± 4.25 | 777.54 ± 25.93 |
| 4. | C4 | 116.54 ± 6.77 | 45.40 ± 3.97 | 666.28 ± 21.10 |
| 5. | C5 | 57.52 ± 4.79 | 53.11 ± 5.34 | 257.33 ± 13.55 |
| 6. | C6 | 72.13 ± 6.10 | 61.82 ± 5.05 | 742.66 ± 23.58 |
| 7. | B1 | 46.75 ± 4.84 | 19.25 ± 3.41 | 271.72 ± 15.03 |
| 8. | B2 | 19.58 ± 3.51 | 16.12 ± 2.80 | 129.30 ± 10.82 |
| 9. | B3 | 19.34 ± 4.61 | 19.23 ± 2.53 | **128.38 ± 10.27** |
| 10. | B4 | 40.90 ± 3.42 | **131.65 ± 6.87** | 1040.9 ± 28.52 |
| 11. | B5 | 51.52 ± 5.04 | 66.33 ± 4.57 | 842.71 ± 25.03 |
| 12. | B6 | 31.81 ± 4.67 | 68.08 ± 4.67 | 772.45 ± 24.18 |
| 13. | S1(W) | 51.67 ± 4.29 | 19.50 ± 2.06 | 283.52 ± 12.50 |
| 14. | S2(W) | **7.33 ± 3.49** | 8.50 ± 2.07 | 257.79 ± 12.61 |
| 15. | S3(W) | 20.23 ± 3.75 | **4.08 ± 1.84** | 146.22 ± 9.58 |
| 16. | S4(W) | 62.67 ± 7.10 | 34.61 ± 19.06 | 762.76 ± 20.51 |
| 17. | S5(W) | 44.24 ± 3.49 | 44.73 ± 3.59 | 584.36 ± 18.61 |
| 18. | S6(R) | 24.34 ± 2.84 | 11.61 ± 1.40 | 153.91 ± 8.51 |
| 19. | S7(R) | 24.85 ± 2.22 | 25.32 ± 2.73 | 672.32 ± 18.99 |
| 20. | S8(R) | 90.08 ± 5.42 | 38.65 ± 3.47 | 667.60 ± 18.87 |
| 21. | P1 | 40.09 ± 7.60 | 38.61 ± 4.49 | 647.01 ± 26.36 |
| 22. | P2 | 97.90 ± 6.46 | 32.69 ± 2.58 | 531.13 ± 16.48 |
| 23. | P3 | 41.44 ± 4.55 | 73.34 ± 5.60 | **1234.5 ± 39.77** |
| 24. | P4 | **157.13 ± 13.03** | 110.42 ± 6.78 | 905.15 ± 31.50 |
| 25. | T1 | 35.35 ± 4.68 | 60.94 ± 4.39 | 696.77 ± 22.56 |
| 26. | T2 | 32.41 ± 4.57 | 38.50 ± 3.85 | 572.34 ± 20.70 |
| 27. | T3 | 45.17 ± 5.09 | 12.62 ± 3.54 | 598.56 ± 21.34 |
| 28. | T4 | 33.67 ± 4.75 | 18.26 ± 3.75 | 707.54 ± 23.14 |
| 29. | T5 | 36.13 ± 2.99 | 33.68 ± 3.54 | 691.96 ± 21.85 |
| 30. | T6 | 58.85 ± 5.49 | 50.06 ± 4.25 | 755.60 ± 24.45 |
| 31. | T7 | 26.50 ± 2.63 | 37.28 ± 3.04 | 626.32 ± 21.14 |
| **Average :** | | **53.06 ± 5.05** | **43.69 ± 4.37** | **590.79 ± 20.43** |

**Table 4:** Comparison of activity concentrations (Bq $kg^{-1}$) in the building materials in different areas of the world.

| **Country** | **$A_{(Ra)}$** | **$A_{(Th)}$** | **$A_{(K)}$** | **References** |
|---|---|---|---|---|
| India | 54 | 65 | 440 | Khandaker et al., 2012 **[31]** |
| Pakistan | 37 | 28 | 200 | Rahman et al., 2013 **[32]** |
| Turkey | 50 | 17 | 246 | Solak et al., 2014 **[33]** |
| China | 68 | 52 | 174 | Xinwei L., 2005 **[34]** |
| Egypt | 134 | 88 | 416 | Ahmed N.K., 2005 **[35]** |
| Italy | 40 | 26 | 244 | Righi S., and Bruzzi, L., 2006 **[36]** |
| Greece | 20 | 13 | 247 | Stoulos S., 2003 **[37]** |
| Bangladesh | 61 | 65 | 952 | Asaduzzaman et al., 2015b **[14]** |
| Present work | **53** | **44** | **591** | - |
| World average | **50** | **50** | **500** | UNSCEAR (1993) **[29]** |

**Table 5:** Radium Equivalent Activity, Absorbed Dose Rate, Annual Effective Dose, External and Internal Hazard Index, Alpha and Gamma Hazard Indices for all investigated building materials.

| Serial no. | Types of Building Materials (no of samples) | Radium Equivalent activity (Bq kg$^{-1}$) | | Absorbed Dose Rates (nGy h$^{-1}$) | | Effective Dose, (mSv y$^{-1}$) | | Hazard Indices | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Range | Mean | $D_{out}$ | $D_{in}$ | $E_{out}$ | $E_{in}$ | $H_{ex}$ | $H_{in}$ | $I_\alpha$ | $I_\gamma$ |
| 1. | Cement (6) | 153 - 340 | 213.87 | 100.75 | 120.90 | 0.12 | 0.72 | 0.58 | 0.81 | 0.42 | 0.78 |
| 2. | Brick (6) | 53 - 309 | 152.29 | 73.25 | 87.90 | 0.09 | 0.52 | 0.41 | 0.51 | 0.18 | 0.56 |
| 3. | White sand (5) | 28– 171 | 98.05 | 46.90 | 56.27 | 0.05 | 0.33 | 0.27 | 0.37 | 0.19 | 0.36 |
| 4 | Red sand (3) | 53 - 197 | 120.79 | 58.01 | 69.61 | 0.07 | 0.41 | 0.33 | 0.45 | 0.23 | 0.45 |
| 5 | Paint (4) | 145-385 | 239.19 | 113.97 | 136.76 | 0.14 | 0.81 | 0.65 | 0.87 | 0.42 | 0.88 |
| 6 | Titles (7) | 109-189 | 140.78 | 68.81 | 82.58 | 0.08 | 0.49 | 0.38 | 0.48 | 0.19 | 0.53 |
| | **Average:** | | **161.02** | **77.10** | **92.52** | **0.10** | **0.55** | **0.43** | **0.58** | **0.27** | **0.59** |
| | **UNSCEAR (2000)** | | **370** | **59** | **84** | **0.07** | **0.41** | **1** | **1** | **1** | **1** |

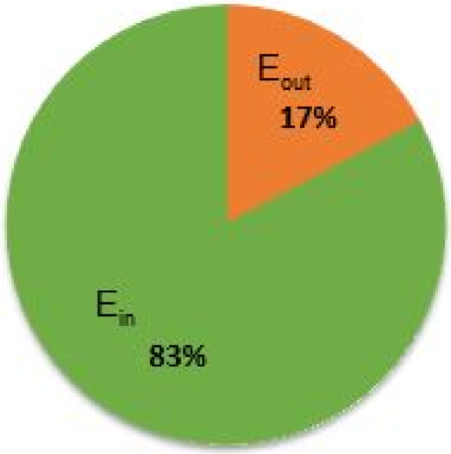

Figure 1: Comparison of average outdoor Annual Effective dose, $E_{out}$ and indoor Annual Effective, $E_{in}$ estimated for building materials in present study.

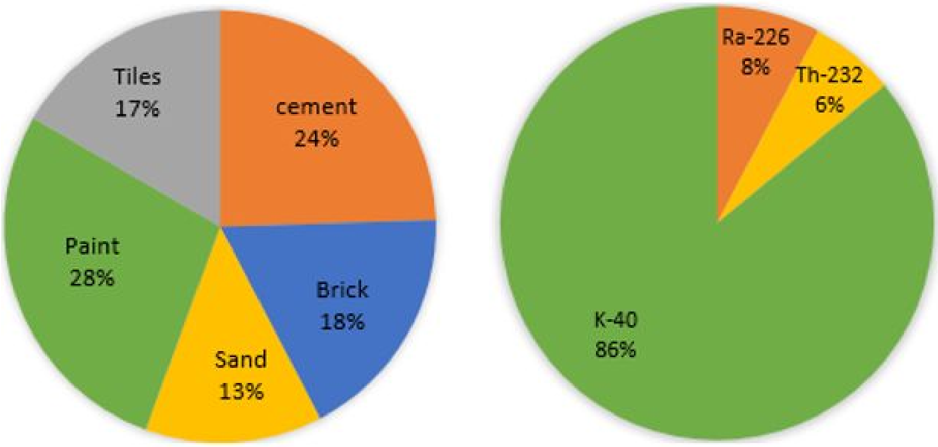

Figure 2: Relative annual effective dose distribution due to $^{226}$Ra, $^{232}$Th, and $^{40}$K contribution in investigated building materials.