



Research Article

Measurement of natural radioactivity and its health hazards associated with the use of different branded cement samples collected from different manufactures in Dhaka city using gamma spectrometry

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ARTICLE INFO

Article History

Received: 27 January 2021

Revised: 29 May 2021

Accepted: 31 May 2021

Keywords: Building materials, Gamma ray spectrometry, High purity germanium detector (HPGe), Activity concentration, Health hazard.

ABSTRACT

The radioactivity of naturally occurring radionuclides ^{226}Ra , ^{232}Th , and ^{40}K was determined in thirteen cement samples collected from different manufactures of Dhaka city. The measurement was performed by gamma-ray spectrometry system using high purity germanium (HPGe) detector. The average activity of ^{226}Ra , ^{232}Th , and ^{40}K in cement samples are found to be $30.23 \pm 15.04 \text{ Bqkg}^{-1}$, $20.10 \pm 11.76 \text{ Bqkg}^{-1}$ and $145.27 \pm 24.14 \text{ Bqkg}^{-1}$, respectively. The average values of radium equivalent activity (R_{eq}), absorbed dose rates (D), internal hazard and external hazard index (H_{ex}), and annual effective dose equivalent are 370 Bqkg^{-1} , 0.27, 0.18, and 27.99 nGyh^{-1} , respectively. The average annual effective dose is found 0.13 mSvy^{-1} , which is less than the recommended value (1 mSvy^{-1}) by the International Commission on Radiological Protection (ICRP-60, 1990), as the maximum permissible annual effective dose to the members of the public. The present study results are discussed and compared with those reported in similar studies and with internationally recommended values. No artificial radioactivity was found in the present study. The results show that the analyzed cement samples do not pose any significant radiation hazard from naturally occurring radionuclides ^{226}Ra , ^{232}Th and ^{40}K reported here and also considered safe for use in buildings construction.

Introduction

We live in an environment where naturally occurring radionuclides happen to exist everywhere with varying concentrations. Among the natural radionuclides, ^{238}U , ^{232}Th , and ^{40}K are the most significance (UNSCEAR, 1988). Radium and its daughter products produce 98.5% of the radiological effects from the uranium series, the contribution from the ^{238}U is replaced by the ^{226}Ra decay product (Cember, 1969). The natural radionuclides emit gamma-

ray, which continuously affects human wellbeing. To estimate the radiation hazard, determination of their concentrations in the surrounding environments of the population is a crucial task. Humans spend most of the time of their life in the buildings, including their houses which are made from different building materials. As a result, the general population is constantly being exposed to ionizing radiation of natural kind resulting from radioactive materials

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come from the Earth's crust, where radionuclides happen to exist in natural quantities (Raghu et al., 2016). The building materials contain trace amounts of radionuclides that are the major source of radiation exposure to humans (Kpeglo et al., 2011). For assessing population exposures, measuring the specific activity of radionuclides in building materials is essential, because the particular activities of the building materials are different from region to region (Sonexay et al., 2018). Building materials also produce significant external and internal dose rates (Florou et al., 1992). The absorbed dose rate in the air from cosmic radiation outdoors is about 30 nGy.h⁻¹ (UNSCEAR, 2000). External exposures to gamma radiation outdoors are mainly generated from terrestrial radionuclides occurring at trace levels in all ground formations. As a result, natural background radiation dependent, mainly on geological and geographical conditions (Florou et al., 2007). In a closed room, the production of ²²²Rn is very high which is responsible for lung cancer. Measuring of natural activity and dose

rate from this building material to implement protective measures when the dose is found to be more than the recommended limits is crucial.

Asaduzzaman et al., found out that the average specific activity of ²³²Th and ⁴⁰K in the cement, brick, and sand samples in Bangladesh were found to be considerably higher than the typical worldwide ranges. In contrast, the levels of ²²⁶Ra in the same samples were within the typical range (Asaduzzaman et al., 2015).

The purpose of this study is to detect natural and artificial radionuclides (²²⁶Ra, ²³²Th, and ⁴⁰K) in cement samples available in the local market of Dhaka city to assess the health risk of the dwellings associated with the investigated cement samples.

Materials and methods

In this study, thirteen cement samples were collected from different manufactures available in the local market of Dhaka city in the year 2017. A list of the collected cement samples was given in Table 1.

Table 1. Location of sample collection with GPS.

Sample Name	Sample ID	Area of Sample Collection	Latitude/Longitude
Shah cement	S-1	Chankharpul	23.7235906/90.3985988
Anwar cement	S-2	Chankharpul	23.7235906/90.3985988
Supercrete cement	S-3	Chankharpul	23.7235906/90.3985988
Tiger cement	S-4	Azimpur	23.7282871/90.380445
Ultratech cement	S-5	Chankharpul	23.7235906/90.3985988
Fresh cement	S-6	Nazira Bazar	23.7195088/90.4062588
Scan cement	S-7	Nazira Bazar	23.7195088/90.4062588
Confidence cement	S-8	Nazira Bazar	23.7195088/90.4062588
Bashundhara cement	S-9	Chankharpul	23.7235906/90.3985988
Crown cement	S-10	Chankharpul	23.7235906/90.3985988
King Brand cement	S-11	Azimpur	23.7282871/90.380445
Deluxe cement	S-12	Nazira Bazar	23.7195088/90.4062588
Premier cement	S-13	Azimpur	23.7282871/90.380445

Sample Collection and Preparation

About (300 gm-400 gm) of the raw sample has been collected. At first, the collected samples were dried properly then all the samples were properly packed and marked for their identification code (IAEA, 1989). The samples were then transported, stored, and processed at the sample preparation laboratory of Atomic Energy Centre, Dhaka (AECDC). Then the samples were transferred to cylindrical plastic containers of 7 cm height and 5.5 cm in diameter and the weights of the samples were recorded using an electrical balance.



Fig. 1. Cement samples after preparation.

The sample-filled plastic containers were sealed tightly with a cap and wrapped with thick vinyl tape around their necks, marked individually with identification number and date of preparation and net weight and then stored for about 30 days to assume secular equilibrium between ^{238}U and ^{232}Th series and their daughter progenies. The photo of the prepared cement sample was shown in Fig.1.

Gamma Spectroscopy and Analysis

HPGe detector is used without any added impurity. It consists of a cylinder-shaped p-type germanium crystal. The samples' detection and measurement of radionuclides were done by gamma spectrometry system,

using a p-type co-axial HPGe detector of 19.6% relative efficiency made by ORTEC. The resolution of the HPGe detector was 2 keV at 1332 keV of the Cobalt-60 gamma-ray line. For analysis of the spectrum of all samples used DSPEC Jr 2.0 (SN16242536) and the MAESTRO software to calculate the concentrations of ^{226}Ra , ^{232}Th , and ^{40}K . All the samples were counted for 5000 seconds. The equal counting time for background and sample measurement was chosen to minimize the uncertainty in the net counts. The spectrum of each sample was analyzed. The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies obtained from the literature. Measured activity concentration of ^{226}Ra and ^{232}Th from their daughter nuclides which consist of daughter nuclides [^{214}Pb (295.2 keV), ^{214}Pb (351.9 keV), ^{214}Bi (609.3 keV), ^{214}Bi (1120.2 keV), ^{214}Bi (1764.4 keV)] and [^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV), ^{228}Ac (911.2 keV), ^{228}Ac (968.9 keV)] respectively were recorded. The activity concentration values quoted assume secular equilibrium for the different isotopic activities in decay chains. Fig. 2 shows the energy spectrum of the counting sample.

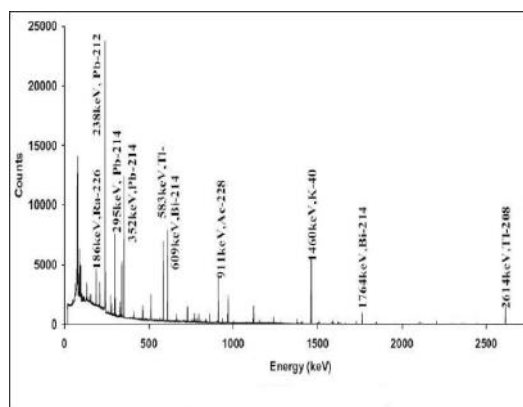


Fig. 2. A typical 'Energy Vs Count' spectrum of samples.

Energy Calibration

The energy calibration of the detector was performed by known activity point source ¹³⁷Cs (661.66 keV) and ⁶⁰Co (1173 keV, 1232 keV).

Efficiency Calibration

The efficiency of a detector is a measure of the number of gamma-rays detected by it from the total number of gamma rays emitted by the source. An accurate efficiency calibration of the spectral analysis to the unit of activity of radionuclides present in a sample. For the low-level activity of the environmental samples, it is desirable to increase the efficiency as much as possible to increase the minimum level of detection (UNSCEAR, 2000). 10 transitions of gamma ray lines of ¹⁵²Eu of known activity with Al₂O₃ matrix radionuclide were used to perform the efficiency calibration. Fig. 3 shown the efficiency calibration graph of the HPGe detector. The efficiency was calculated using the following formula:

$$\text{Efficiency} = \text{CPS}/(\text{DPS} \times I_{\gamma})$$

Where, CPS = counts per second for the radionuclide present in the standard sample,

DPS = disintegration per second, and

I_γ = γ -ray intensity of the source.

The detected count/sec can be measured as,

$$\text{CPS} = \frac{\text{Net area under a photo peak}}{\text{Counting time (in sec)}}$$

Calculation of Activity Concentration

The activity concentration of each cement sample was determined by using the calibrated HPGe detector. The radioactivity of natural radionuclides, namely, uranium and thorium series, as well as ⁴⁰K, was investigated in cement samples collected from the local market of Dhaka city.

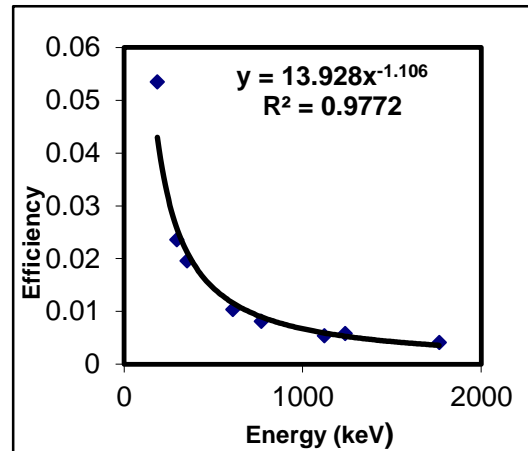


Fig. 3. Efficiency calibration graph.

The activity concentration of each radionuclide in the cement sample was measured by using the net count rates (cps) for the same counting time under the selected photo peaks, the weight of the sample, photopeak efficiency, and the gamma intensity at a specific energy as given by the following equation (Knoll, 1998):

$$A = \frac{N \times 100 \times 1000}{\epsilon \times P_{\gamma} \times w (gm)} \dots \dots \dots (1)$$

Where,

A= activity of the sample in BqKg⁻¹

N = Net counts per second (c. p. s) = (Sample c. p. s) – (Background c. p. s)

ε= Efficiency in percent

P_γ= Transition probability of gamma-ray

w = Weight of the sample in gm

Error in Radioactivity Measurement

The percentage of sample counting error for the radioactivity measurement is found with the help of the following relation 2 given below (Monira et al., 2005).

The errors in the measurement have been expressed in terms of standard deviation ($\pm\sigma$), where σ is expressed as,

$$\sigma = \left[N_s/T_s^2 + N_b/T_b^2 \right]^{1/2} \dots\dots\dots(2)$$

Where,

N_s is the counts measured in time T_s , and N_b is the background counts measured in time T_b . The standard deviation ($\pm\sigma$) in cps was converted into activity in $BqKg^{-1}$.

Calculation of Radium Equivalent Activity (Ra_{eq})

The inhomogeneous distribution of naturally occurring radionuclides is due to the disequilibrium between ^{226}Ra and its decay products. For uniformity in exposure estimates, the radionuclide concentrations are defined in terms of ‘Radium equivalent activity’ (Ra_{eq}) in $Bqkg^{-1}$. This allows comparison of the specific activity of materials containing different amounts of ^{226}Ra , ^{232}Th and ^{40}K according to Beretka and Mathew (Beretka, 1985) as follows:

$$Ra_{eq} (Bqkg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \dots\dots\dots(3)$$

Where, C_{Ra} , C_{Th} , and C_K are the specific activities of ^{226}Ra , ^{232}Th , and ^{40}K , respectively, in $Bqkg^{-1}$.

Calculation of Absorbed Dose Rate

The absorbed dose rate in air at height of 1.0 meter above the ground for ^{226}Ra , ^{232}Th and ^{40}K , assuming a uniform dose distribution, was estimated using the formula given below (Beck and de Planque, 1968),

$$D = 0.52813C_{Th} + 0.38919C_{Ra} + 0.03861C_K \dots\dots\dots(4)$$

Where, C_{Ra} , C_{Th} and C_K are activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in $Bqkg^{-1}$ and D in $nGyh^{-1}$ respectively.

The absorbed dose rate in the air at the height of 1 meter above the ground for ^{226}Ra , ^{232}Th , and ^{40}K , assuming a uniform dose distribution,

was estimated using the formula given below (Beck and de Planque, 1968),

$$D = 0.52813C_{Th} + 0.38919C_{Ra} + 0.03861C_K \dots\dots\dots(4)$$

Where, C_{Ra} , C_{Th} , and C_K , are activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in $Bqkg^{-1}$, and D in $nGyh^{-1}$, respectively.

Calculation of External and Internal Hazard Indices

Internal hazard index (H_{in}) is used for consideration the internal radiation from radon ^{222}Rn and its daughter in building material. The (H_{in}) calculated by (Lu and Zhang, 2006):

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \dots\dots\dots(5)$$

On the other hand, external gamma radiation (H_{ex}) is dose product by building material radionuclides. The (H_{ex}) equivalent upper limit of $1mSvy^{-1}$ and calculated by (Al-Hamarneh and Awadallah, 2009):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \dots\dots\dots(6)$$

Where C_{Ra} , C_{Th} , and C_K are the activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in $Bqkg^{-1}$. The value of H_{ex} must be lower than unity to keep the radiation hazard insignificant.

Calculation of Annual Effective Dose

The annual effective dose, D_E due to gamma radiation from building materials with the annual exposure time of 7000h, was calculated as follows, $D_E = 0.7(Sv.Gy^{-1}) \times 7000(h) \times 10^{-6} \times D(nGyh^{-1}) \dots\dots\dots(7)$

Results and discussion

The radium equivalent activity, dose rate, external and the internal hazards, and annual effective dose were calculated to assess the health risk associated with the cement samples.

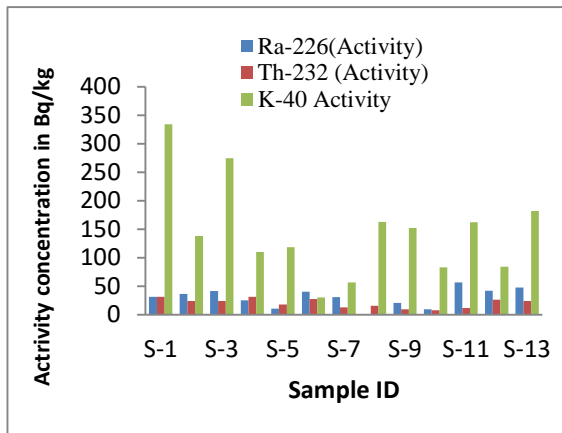


Fig. 4. The variations of activity of ²²⁶Ra, ²³²Th, ⁴⁰K in cement samples.

The activity of ²²⁶Ra, ²³²Th, ⁴⁰K in cement samples is shown in Fig. 4. The activity of ²²⁶Ra in cement samples was found to be varied from 0.88 ± 12.75 Bqkg⁻¹ in S-8 (Confidence cement) to 56.52 ± 14.88 Bqkg⁻¹ in S-11 (King Brand cement) with an average value of 30.23 ± 15.04 Bqkg⁻¹. The activity concentration of ²³²Th in cement samples was found to be varied from 9.45 ± 5.64 in S-9 (Bashundhara cement) to 31.49 ± 17.39 Bqkg⁻¹ in S-1 (Shah cement) with an average value of 20.10 ± 8.76 Bqkg⁻¹. The activity concentration of ⁴⁰K in cement samples was found to be varied from 82.77 ± 11.06 Bqkg⁻¹ in S-10 (Crown cement) to 334.22 ± 44.33 Bqkg⁻¹ in S-1 (Shah cement) with an average value 145.27 ± 24.14 of Bqkg⁻¹. The variation of the radioactivity of the cement is due to the varying amounts of uranium, thorium, and potassium contents in the different geological materials. According to UNSCEAR, the permissible world average concentration of radium, thorium, and

potassium are about 35 Bqkg⁻¹, 30 Bqkg⁻¹, 400 Bqkg⁻¹, respectively (UNSCEAR, 2000). The average activity concentration of the radium, thorium, and potassium of thirteen cement samples is below the permissible world.

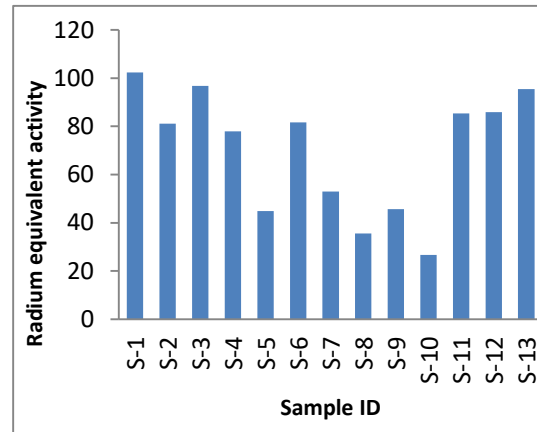


Fig. 5. The variation of radium equivalent activity (Ra_{eq}) of the cement sample.

Radiological indices

The index values of radiological parameters such as radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}), external hazard index (H_{ex}), absorbed dose rate (D), and annual effective dose rate (D_E) has been calculated to estimate the radiological risk due to the presence of ²²⁶Ra, ²³²Th, and ⁴⁰K in investigated cement samples. Fig. 5 shows the variation of radium equivalent activity (Ra_{eq}) of the cement sample. Radium equivalent activity (Ra_{eq}) values ranged from 26.68 Bqkg⁻¹ in S-10 to 102.32 Bqkg⁻¹ S-1 with an average of 70.17 Bqkg⁻¹. The world average permissible limit for radium equivalent activity is 370 Bqkg⁻¹ (UNSCEAR, 2000). The calculated average value of radium equivalent activity in all samples was below the average world value.

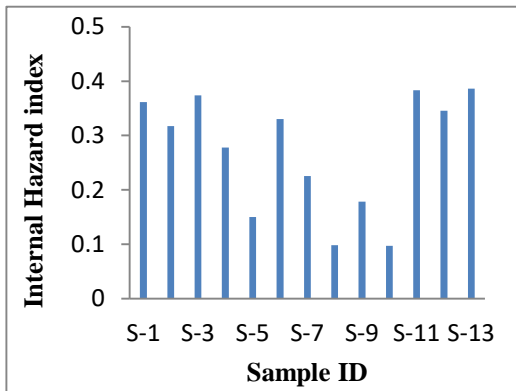


Fig. 6. Internal hazard index (H_{in}). The internal hazard index (H_{in}) was ranged from 0.09 to 0.38, with an average value of 0.27.

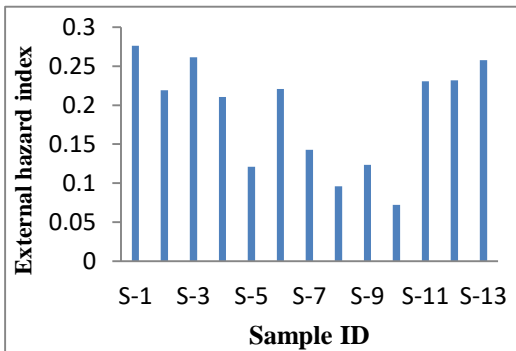


Fig. 7. External hazard index (H_{ex}). The external hazard index (H_{ex}) was ranged from 0.07 to 0.28, with an average value of 0.189.

Fig. 6 and Fig.7 show that the mean values of H_{ex} and H_{in} are both below the recommended level of 1.

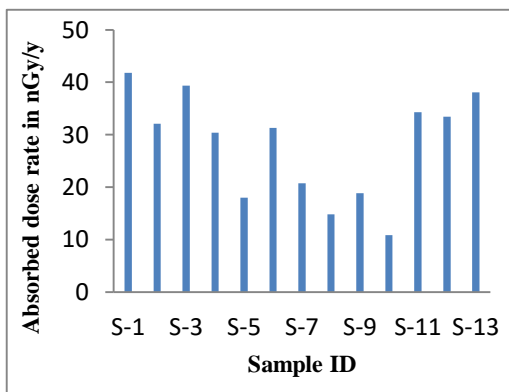


Fig. 8. The variation of absorbed dose rate in nGy/h.

Fig. 8 shows the variation of absorbed dose rate in nGy/h. The lowest and highest absorbed dose rate (D) was found 10.88 nGyh⁻¹ in S-10 to 41.82 nGyh⁻¹ in S-1 with an average of 27.99 nGyh⁻¹.

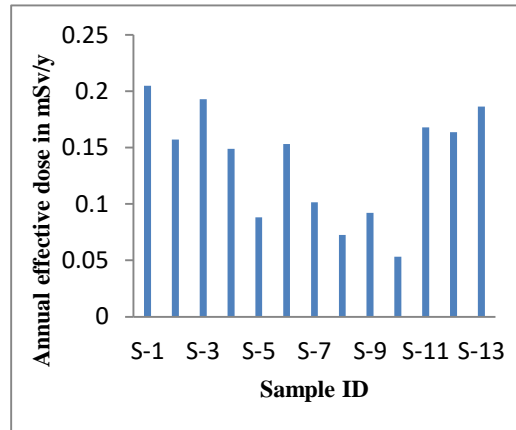


Fig. 9. The variation of annual effective dose in mSv/y.

The permissible world average value for absorbed dose rate is 55 nGyh⁻¹ (UNSCEAR, 2000). The calculated average value of external hazard index in all samples was below than the world average. Fig. 9 shows the total annual effective dose (D_E) from the cement sample. The highest and lowest annual effective dose was found 0.20 mSvy⁻¹ in S-1 (Shah cement) to 0.05 mSvy⁻¹ in S-10 (Crown cement) with an average of 0.137 mSvy⁻¹. The permissible world average value of annual effective dose is 1 mSvy⁻¹ (UNSCEAR, 2000). The calculated average value of the annual effective dose rate in all samples was below the average world value.

From table 2, it is clear that the result of the present study is below the value of India, Pakistan, China, Turkey, Tanzania but high than Nigeria, Saudi Arabia Tehran. The results show considerable variations in the measured

Table 2. Comparison of the activity concentrations of natural radionuclides in cement samples in different countries of the world.

Country	Building materials type	Average activity (Bqkg ⁻¹)			References
		²²⁶ Ra	²³² Th	⁴⁰ K	
India	Cement	69.95	53.19	318.62	(Raghu et al., 2016)
Bangladesh	Cement	40.80	63.00	955.90	(Roy et al., 2005)
Bangladesh	Cement	60.50	64.70	952.20	(Asaduzzaman et al., 2015)
Bangladesh	Cement	120.20	132.40	505.70	(Mollah et al., 1986)
Nigeria	Cement	2.16	7.82	114.30	(Ademola et al., 2017)
Pakistan	Cement	26.70	28.60	273.00	(Khan et al., 2001)
Pakistan	Cement	34.20	29.10	295.10	(Aslam, 2012)
China	Cement	56.50	36.50	173.20	(Xinwei, 2005)
Saudi Arabia	Cement	18.93	15.76	114.80	(Alkhomashi et al., 2017)
Turkey	Cement	40.50	26.10	267.10	(Turhan, 2008)
Tehran	Cement	17.00	9.00	422.00	(Amiri et al., 2014)
Tanzania	Cement	46.00	28.00	228.00	(Amasi et al., 2014)
World average value	Cement	35.00	30.00	400.00	(UNSCEAR, 2000)
Bangladesh	Cement	30.23	20.10	145.27	Present Work

values of samples of building material originating from different areas. One of the former study (Mollah et al., 1986) has a higher value than the present study, could be the ingredients of the raw materials of cement samples and other materials like fly ash.

Conclusions

Thirteen cement samples collected from local markets of Dhaka city have been analyzed by gamma ray spectrometry system using high purity germanium detector. In this present study, the mean activity of ²²⁶Ra, ²³²Th, and ⁴⁰K in cement samples was found at 30.23±15.04 Bqkg⁻¹, 20.10±11.76 Bqkg⁻¹, and 145.27±24.14 Bqkg⁻¹, respectively.

The calculated average value of absorbed dose rate is found lower than the world average limit 55 nGyh⁻¹. The average annual effective dose rate is found at 0.13 mSvy⁻¹, which is less than the recommended level of 1.0 mSvy⁻¹. No artificial radionuclides were found in these samples.

Therefore, the investigated cement samples are considered safe for constructing buildings in the study area and do not pose any significant source of radiation hazard. The results found in the present study cover only Dhaka city's local market. To develop a countrywide baseline of natural radioactivity, particularly, ²²⁶Ra where indoor ²²²Rn builds up, to expanding the program cover the whole country is necessary.

Acknowledgement

The authors are grateful to the scientists and all the staff of the Health Physics Division, Atomic Energy Centre Dhaka, Bangladesh Atomic Energy Commission for their kind assistance and for providing laboratory facilities.

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