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^{232}Th, ^{226}Ra, AND ^{40}K ACTIVITIES AND ASSOCIATED RADIOLOGICAL HAZARDS IN BUILDING MATERIALS OF ISLAMABAD CAPITAL TERRITORY, PAKISTAN

by

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Radioactivity levels in building materials, collected from the Islamabad capital territory have been determined by using a gamma spectrometric technique. Measured specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in material samples ranged from 8   1 to 116   6 Bq/kg, 9   1 to 152   5 Bq/kg, and 29   6 to 974   23 Bq/kg, respectively. The radium equivalent activity, absorbed dose rate, annual effective dose, and gamma index were evaluated from the measured amounts of radioactivity to assess the radiation hazard associated with the studied building materials. The mean radium equivalent activity, the absorbed dose rate and annual effective dose estimated ranged from 81   6 to 221   11 Bq/kg, 38   3 to 104   5 nGy/h, and 0.23   0.02 to 0.64   0.03 mSv, respectively. The ranges of the calculated Ra_{eq} were found to be lower than the values recommended for construction materials (370 Bq/kg). The mean values of the internal and external hazard indices were found in the range of 0.30   0.02 to 0.78   0.05 and 0.22   0.02 to 0.60   0.03, respectively. The results of the materials examined indicate no significant radiological hazards arise from using such material in building construction.

Key words: building material, radioactivity, gamma spectrometry, radiological hazard, annual effective dose

INTRODUCTION

Radioactive nuclides have always been present in the natural environment and studies of the background radiation levels are of great importance being considered as one of the main sources of exposure to human being. Most of the environmental radiation contribution comes from the radionuclides which are members of the natural radioactive series and ^{40}K [1-3]. Beside medical exposures and cosmic radiation, building materials containing naturally occurring radionuclides are the main source of exposure. The naturally occurring radionuclides in building materials contribute to radiation exposure in two ways: the external radiation originating from gamma radiation of the ^{238}U and ^{232}Th decay series and of ^{40}K and the internal radiation due to radon inhalation, leading to deposition of its decay products in the respiratory tract. Since all building materials contain natural radioactive isotopes, the exposure of population to ionizing radiation originates from the walls of the building in which people live [4-8]. The knowledge of the natural radioactivity in these materials is important for determination of the amount of public radiation exposure because people spend most of their time indoors.

During the last three decades considerable attention has been devoted to the exposure originating from naturally occurring radionuclides, particularly to the control of natural radiation in building materials. Building materials are one of the sources which cause direct radiation exposure and generally, dose contributions to residents in dwellings from building materials are small as compared to those from underlying bedrock and soil. However, many research articles available in open publications have identified building materials as the major radiation contributors [9-15]. The content of ^{226}Ra and ^{232}Th in these materials can also increase the concentrations of ^{222}Rn and ^{220}Rn and their daughters in a building. It is well known that sustained exposure of humans to a substantial concentration of indoor radon and its short lived progeny is a cause of lung cancer [16, 17].

There are a few data available on the radioactivity levels of building materials used in Pakistan. Therefore, there is a need to investigate the radioactivity levels of building materials such as sand, bricks, cement, marble, gravel, and ceramics used in building constructions. The latter would contribute to the estab-

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lishment of a baseline map of the environmental radioactivity level in Pakistan. The measurement of the radioactivity concentrations of building materials is important because of the living environment. If the building materials contain high levels of radioactivity, then the radiological hazards on people may be significant. To limit the exposure of the population to ionizing radiation, there is a need to control and minimize the materials used in construction having higher contents of radioactivity.

The objective of the present study is to examine the specific activity of ^{232}Th, ^{226}Ra, and ^{40}K in some local building materials used commonly in Pakistani dwellings. Knowledge of radioactivity levels is useful in setting the standards and national guidelines in light of the international recommendations and in assessing the associated radiological hazard. Furthermore, measurement of natural radioactivity due to gamma rays from these materials helps to implement precautionary measures whenever the levels are found to be above the recommended limits. In addition, the results of present study are compared with results of local studies and those of other countries of the world.

MATERIALS AND METHODS

Sample collection and preparation

Six types of commonly used building materials such as sand, red bricks, cement, marble, gravel, and ceramics were collected from different construction sites and suppliers of the studied area. Ten samples were obtained for each type of building material and total 60 samples were prepared for gamma-ray spectrometry. The samples were then heated in an electric oven at 110 °C up to 48 hours in order to remove moisture contents and to attain constant sample weight. After drying, the samples were crushed, ground, and passed through a sieve of 2 mm mesh size. The homogenized samples were packed in Marinelli beakers with the same geometry as that of the reference materials. The beakers were hermetically sealed to avoid any leakage of radon gas and kept for a period of 30 days to establish secular equilibrium among the progenies of ^{238}U and ^{232}Th decay series. Approximately seven half-lives of ^{222}Rn, are generally considered sufficient for the establishment of minimal secular equilibrium among the progenies of ^{238}U and ^{232}Th decay series. These prepared samples are used for gamma counting.

GAMMA SPECTROMETRY

Activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K isotope in 60 representative samples of building materials were measured using high resolution gamma spectrometry system. A high purity vertical HPGe detector with an electronics card and a PC was used for the measurement of γ-rays emitted from the samples. The spectrometry system comprised of high-purity germanium coaxial detector having a relative efficiency of 30% with respect to NaI(Tl) detector and an active volume of 180 cm^3. The energy resolution of the detector was 2.0 keV (the full width at half- maximum) at 1332 keV γ-ray line of ^{60}Co. The detector was housed in a 15 cm thick cylindrical lead shield in order to effectively reduce the natural background. The equipment was calibrated against reference sample with known activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K. Prior to sample measurement, the environmental γ-ray background was determined with empty Marinelli beakers under the identical measurement conditions as the active samples measured [1, 7, 18].

Samples were counted for 6.5 10^3 s. The data acquisition was carried out using commercially available computer software Genie-2000. For a nuclide having more than one peak in the spectrum the concentration was derived by arithmetic mean of activities obtained from several different peaks in the spectrum. Peaks from ^{214}Bi and ^{214}Pb at 351.9 and 609.3 keV were used to represent the activity of ^{238}U. For confirmation, the other lines (1120, 1764, and 2204 keV) were also monitored. To measure the activity of ^{232}Th, the peaks of ^{212}Bi (727.33 keV), ^{228}Ac (209.25, 409.46, 463.0, 794.95, 911.20, 964.77, 968.97 keV) and ^{212}Pb (238.63, 300.09 keV) were used. Activity of ^{40}K was determined directly through its gamma photo peak at 1460.82 keV [1, 7, 18].

RESULTS AND DISCUSSION

Radioactivities of ^{226}Ra, ^{232}Th, and ^{40}K measured in different building materials used for private and public buildings are given in tab. 1. It can be seen from these results that the lowest mean value of ^{226}Ra concentration is 8 1 Bq/kg measured in marble, while the highest mean value for the same radionuclide is 116 6 Bq/kg recorded in ceramics. The lowest activity of ^{232}Th is 9 1 Bq/kg recorded in cement and the highest value is 152 5 Bq/kg measured in ceramics. The measured activity values of ^{40}K vary from 29 6 Bq/kg in marble to 974 23 Bq/kg in gravel. It can be seen from tab. 1 that ^{40}K almost always contributes to the most specific activity compared to ^{238}U and ^{232}Th. The average concentration of ^{232}Th, ^{226}Ra, and ^{40}K radionuclides in the studied building materials are 46 3, 49 3, and 355 16 Bq/kg, respectively. The mean values of the three radionuclides in all samples under investigation are shown in fig. 1.

RADIUM EQUIVALENT ACTIVITY

Thorium, radium, and potassium are not uniformly distributed in soil or rocks, from which build-

Table 1. Measured activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K in all samples collected from Islamabad capital territory, Pakistan

Sample tape	^{232}Th [Bqkg^{-1}]			^{226}Ra [Bqkg^{-1}]			^{40}K [Bqkg^{-1}]		
	Max.	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean
Sand	63 ± 4	17 ± 2	37 ± 3	38 ± 3	15 ± 1	26 ± 2	621 ± 18	105 ± 13	416 ± 16
Brick	123 ± 5	39 ± 2	77 ± 4	92 ± 5	27 ± 2	67 ± 4	893 ± 21	141 ± 15	572 ± 18
Cement	49 ± 5	09 ± 1	28 ± 2	73 ± 5	17 ± 2	39 ± 3	322 ± 21	63 ± 13	160 ± 16
Marble	45 ± 3	12 ± 1	27 ± 2	65 ± 4	8 ± 1	29 ± 2	565 ± 21	29 ± 6	172 ± 14
Gravel	81 ± 5	17 ± 1	46 ± 3	76 ± 5	14 ± 1	44 ± 3	974 ± 23	87 ± 15	423 ± 18
Ceramics	152 ± 5	19 ± 3	78 ± 4	116 ± 6	39 ± 3	74 ± 5	689 ± 22	92 ± 11	386 ± 16

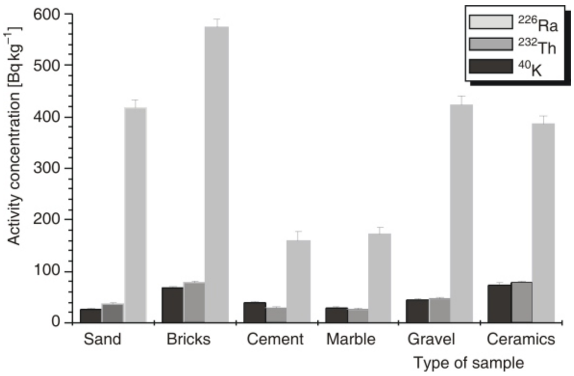

Figure 1. Mean values of ^{232}Th, ^{222}Ra, and ^{40}K activities [Bqkg^{-1}] in the listed building samples collected from Islamabad capital territory, Pakistan

ing materials are derived. Uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of materials containing different amounts of ^{232}Th, ^{226}Ra, and ^{40}K. It is calculated by the following expression [10, 19].

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (1)$$

where C_{Th}, C_{Ra}, and C_K are the activity in Bq/kg of ^{232}Th, ^{226}Ra, and ^{40}K, respectively. Equation (1) is based on the fact that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma ray dose rate [2, 5]. Table 2 summarizes the Ra_{eq} results in all samples under investigation.

It is evident from the results that the lowest mean value of Ra_{eq} is 81 6 Bq/kg calculated in marble, while the highest value is 221 11 Bq/kg calculated in bricks. The overall mean value of the Ra_{eq} for all of the studied materials found to be 144 9 Bq/kg. From these results it is obvious that the Ra_{eq} varies considerably in the different materials and also within the same type of material collected from different areas. The large variations in radium equivalent activities also indicate that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material. In the present work, the calculated Ra_{eq} values of all the materials are less than the maximum recommended value to be used for homes (370 Bq/kg) [20]. The obtained results are in a reasonable accord with other published data given in tab. 3.

ABSORBED GAMMA DOSE RATE IN AIR

It is an established fact that some of the buildings may cause excessive radiation doses to the human

Table 2. Calculate values of radium equivalent activity, absorbed dose rate, annual effective dose equivalent, extrernal and internal hazard indices for all samples

Sample type		Sand	Brick	Cement	Marble	Gravel	Ceramics
Ra_{eq} [Bqkg^{-1}]	Max.	136 8	275 12	144 12	118 7	191 11	333 16
	Min.	88 6	188 9	55 6	41 4	80 6	90 9
	Mean	110 7	221 11	91 8	81 6	142 9	216 12
Absorbed dose rate [nGyh^{-1}]	Max.	73 4	130 6	66 6	57 4	95 5	155 7
	Min.	44 3	88 4	25 3	19 2	38 3	42 4
	Mean	53 3	104 5	42 4	38 3	67 4	100 6
Annual effective dose [mSv]	Max.	0.45 0.03	0.80 0.04	0.41 0.04	0.35 0.02	0.58 0.03	0.95 0.04
	Min.	0.27 0.02	0.54 0.03	0.16 0.02	0.12 0.01	0.23 0.02	0.26 0.02
	Mean	0.32 0.02	0.64 0.03	0.26 0.02	0.23 0.02	0.41 0.03	0.61 0.03
Internal hazard index	Max.	0.49 0.03	0.93 0.05	0.54 0.04	0.46 0.03	0.62 0.04	1.21 0.06
	Min.	0.29 0.02	0.68 0.04	0.24 0.02	0.14 0.01	0.29 0.02	0.35 0.03
	Mean	0.37 0.02	0.78 0.04	0.35 0.03	0.30 0.02	0.50 0.03	0.78 0.05
External hazard index	Max.	0.41 0.02	0.74 0.03	0.39 0.03	0.32 0.02	0.52 0.03	0.90 0.04
	Min.	0.24 0.02	0.51 0.02	0.15 0.02	0.11 0.01	0.22 0.02	0.24 0.02
	Mean	0.30 0.02	0.60 0.03	0.25 0.02	0.22 0.02	0.38 0.02	0.58 0.03

Table 3. Comparison of radium equivalent activities in the listed building materials used in Islamabad capital territory, Pakistan with those of other countries

Country	Sand [Bqkg^{-1}]	Bricks [Bqkg^{-1}]	Cement [Bqkg^{-1}]	Marble [Bqkg^{-1}]	Gravel [Bqkg^{-1}]	Ceramics [Bqkg^{-1}]	Reference
Australia	70	883	115		115		[10]
Zambia	117 12	180 22	79 11		24		[19]
China	96.4 2.3	178.3 6.2	162.8 6.3		82.6 4.6		[22]
Germany	59	640	70		322		[23]
Egypt	16.6	77	292 130	436 199	19.7	252 118	[9, 25]
Jordan	82		78.5	42.9			[11, 26]
India	84.15	69.15	108.5	60		121.16	[27]
Algeria	28 7.1	190 9.5	112 8.1	73 4.1	58		[27]
Bangladesh	87.5 38.0	127.1 9.85	172.8 19.8		121.3 22.6		[28]
Kuwait	45.4	41.6	45.1	4.2			[29]
Malaysia	136 33	895 107	188 27				[30]
Syria	130	105	47				[31]
Turkey	101.2 50.6	144.0 28.2	101.9 31.3	9.8 3.4	95.6 14.2	218.3 56.8	[32]
Cameroon	104.6	193.34	70.1	10.15	312.51	36.03	[33]
Hong Kong	70	213	55				[34]
Cuba	53.5	134.6	73.9		45.2		[35]
Brazil	34.0	247.7 170.3	50.9		82		[36]
Pakistan	129 54	186	68 9	67 60			[37, 38]
Present study	110 7	221 11	91 8	81 6	142 9	216 12	

body due to gamma rays emitted by the progenies of ^{226}Ra (*i. e.* ^{214}Pb and ^{214}Bi). Daughter products of ^{232}Th decay chain and ^{40}K also contribute to the total body radiation dose. The air absorbed dose rate in units of nGy/h can be defined if the radionuclide concentrations of ^{232}Th, ^{238}U, and ^{40}K in Bq/kg are known. It was calculated using the formula proposed by UNSCEAR, 2000 [17].

$$D = 0.427C_U + 0.662C_{Th} + 0.0432C_K \qquad (2)$$

where C_U, C_{Th}, and C_K are the activity concentrations of ^{232}Th, ^{238}U, and ^{40}K respectively in building material samples. In the above conversions, it is assumed that all the decay products of ^{232}Th and ^{226}Ra are in radioactive equilibrium with their precursors. Equation (2) is used to calculate the absorbed dose rate in air at 1.0 m above the ground from the measured radionuclide concentration in building materials for general public of the studied area. Results of the dose rates in air from investigated building materials are summarized in tab. 2. It is obvious from these results that the lowest value of the dose rate 38 3 nGy/h found in marble samples, whereas the highest value is found in bricks samples (104 5 nGy/h). The overall mean dose rate of the studied building material was 67 4 nGy/h.

ANNUAL EFFECTIVE DOSE

The annual effective dose equivalent expected to be received by the occupants from the walls of dwelling rooms can be calculated using the following relation

$$E = (\text{Absorbed dose})\,\text{nGy/h} \quad 8760\,\text{h} \quad 0.7\ \text{Sv/Gy} \qquad (3)$$

The annual effective dose equivalent was calculated using a conversion factor of 0.7 Sv/Gy, which converts absorbed dose in air to human effective dose in adults. The values obtained range from 0.23 0.02 to 0.64 ± 0.03 mSv with an average value of 0.41 0.03 mSv. It can be seen from tab. 2 that annual effective dose values are in agreement with the corresponding worldwide values given in UNSCEAR-2000 [17]. It may be concluded from the overall results in this study that building materials used in Islamabad capital territory for building construction do not pose a radiological hazard.

EXTERNAL HAZARD INDEX

To calculate the external γ-radiation dose from building materials, the following expression is used to define the external hazard index (H_{ex})

$$H_{ex} \quad \frac{A_{Th}}{259} \quad \frac{A_{Ra}}{370} \quad \frac{A_K}{4810} \qquad (4)$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bq/kg for the material. The value of this index must be less than unity for the radiation hazard to be negligible. The obtained values of H_{ex} are shown in tab. 2. The results show that the minimum mean value 0.22 0.02 obtained for marble while the maximum value 0.60 0.03 for bricks sample with an overall mean value 0.39 0.02 of all investigated materials. It is evident from these results; the studied materials are radiologically safe and can be used in building construction [21].

INTERNAL HAZARD INDEX

Internal hazard index is calculated to determine the radiation hazard to respiratory organs due to radon and its short lived daughter products. It is quantified by the following expression [22, 23]

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

where $A_{\rm Th}$, $A_{\rm Ra}$, and $A_{\rm K}$ are the activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K, respectively, in Bq/kg for the material. For the safe use of a material in the construction of dwellings $H_{\rm in}$ should be less than unity [24]. The values of $H_{\rm in}$ for samples under investigation are shown in tab. 2. It is evident from the table, $H_{\rm in}$ values range from 0.30 0.02 to 0.78 0.04 for marble and bricks, respectively. The mean value of $H_{\rm in}$ found to be 0.51 0.03 for all the materials. From these results, it is obvious that all the calculated $H_{\rm in}$ values are less than unity, therefore, the studied materials are safe to be used in construction of dwellings and workplaces.

CONCLUSIONS

The activity concentrations of building materials used in Islamabad capital territory were determined using gamma-ray spectrometry. The measured mean radium concentrations in sand and marble are among the lowest measured values while the maximum values were found in bricks and ceramics. The activity concentrations measured in ceramics and bricks were the maximum compared to those of other building materials for ^{232}Th and ^{40}K, respectively. The radium equivalent concentrations in cement and marble are among the lowest measured values while the maximum values were found in bricks. The overall mean radium equivalent values found to be 144 9 Bq/kg for all the studied building materials which is well below the defined limit of 370 Bq/kg. The mean external and internal hazard indices of the selected building materials were found to be 0.39 and 0.51, respectively. The mean estimated dose rate was 67 nGy/h whereas mean annual effective dose equivalent found to be 0.41 mSv per year in this study for all the evaluated building materials. Thus, the studied materials do not pose a significant radiological hazard when used in construction of dwellings or workplaces.

AUTHOR CONTRIBUTIONS

Experiment was carried out by S. U. Rahman and M. Rafique analyzed and discussed the results. The manuscript was written by S. U. Rahman and M. Rafique, and the figures were prepared by M. Rafique.

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АКТИВНОСТ ^{232}Th, ^{226}Ra и ^{40}K И ОДГОВАРАЈУЋИ РАДИОЛОШКИ РИЗИК ГРАЂЕВИНСКОГ МАТЕРИЈАЛА ИЗ ОБЛАСТИ ГРАДА ИСЛАМАБАДА У ПАКИСТАНУ

Гама спектрометријским мерењима одређени су нивои радиоактивности у грађевинским материјалима прикупљеним у области града Исламабада. Измерене специфичне активности у узорцима материјала биле су у опсегу од 8   1 Bq/kg до 116   6 Bq/kg за ^{226}Ra, 9   1 Bq/kg до 152   5 Bq/kg за ^{232}Th и од 29   6 Bq/kg до 974   23 Bq/kg за ^{40}K. На основу измерене радиоактивности, одређени су еквивалентна активност радијума, јачина апсорбоване дозе, годишња ефективна доза и гама индекс ради процене радијационог ризика ових грађевинских материјала. Добијене су вредности у следећим опсезима: за средњу еквивалентну активност радијума од 81   6 Bq/kg до 221   11 Bq/kg, за јачину апсорбоване дозе од 38   3 nGy/h до 104   5 nGy/h и за годишњу ефективну дозу од 0.23   0.02 mSv до 0.64   0.03 mSv. Вредности израчунате еквивалентне активности радијума биле су ниже од препоручених вредности за грађевинске материјале (370 Bq/kg). Средње вредности индекса унутрашњег и спољашњег ризика износиле су од 0.30   0.02 до 0.78   0.05 и од 0.22   0.02 до 0.60   0.03, респективно. Није показана значајна радиолошка опасност од коришћења ових грађевинских материјала.

Кључне речи: грађевински материјал, радиоактивност, гама спектрометрија, радиолошки ризик