

RADIOACTIVITY MEASUREMENT OF PRIMORDIAL RADIONUCLIDES IN AND DOSE EVALUATION FROM MARBLE AND GLAZED TILES USED AS COVERING BUILDING MATERIALS IN TURKEY

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Measurements of the natural radioactivity arising from primordial radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in marble and glazed tile samples used covering building materials in Turkey were carried out by gamma-ray spectrometer with a high purity germanium detector. The mean activity concentrations of the ^{226}Ra , ^{232}Th and ^{40}K in marble and glazed tile samples were found as 8.2, 5.5 and 58.1 Bq kg⁻¹ and 81.2, 65.4 and 450.1 Bq kg⁻¹, respectively. The radiation doses received by occupants of buildings in which the sample marble and glazed tiles might be used are estimated using measured activity concentrations of constituent primordial radionuclides and dose conversion factors evaluated by the European Commission from models of tile use. Results obtained are presented for each radionuclide, analysed and compared with relevant national and international legislation, guidance and report, and with the results obtained from other studies. Results show that the use of such decorative building materials in the construction of domestic homes or workplaces in Turkey is unlikely to lead to any significant radiation exposure to the occupants.

INTRODUCTION

Covering or decorative building materials derived from rock and soil include primordial radionuclides such as uranium–radium (^{238}U – ^{226}Ra) and thorium ^{232}Th series, and potassium radioisotope (^{40}K) varying from one country to another and from one location to another in the same country⁽¹⁾. Radiation emitted from these radionuclides in the covering building materials like other building materials are sources of external and internal exposure indoors. External radiation exposure is caused by direct gamma radiation from the members of the ^{226}Ra and ^{232}Th series along with ^{40}K , while internal radiation exposure is due to the inhalation of the radioactive inert gas radon (^{222}Rn) and thoron (^{220}Rn) and their short-lived decay products, which are exhaled from building materials indoors. The precursors of ^{226}Ra in the ^{238}U series are generally ignored because 98.5 % of the radiological effects of the ^{238}U series are produced by the ^{226}Ra and its daughter products^(2, 3). Determination of the radioactivity levels of building materials used in construction sectors is crucial in the evaluation of possible radiological hazards to human health because exposures of the occupants to the radiation can be increase discernibly by the usage of building materials including above the normal level of the primordial radionuclides. Hence, remarkable efforts have been made to determine the radioactivity in

various building materials used in the different countries^(4–10).

In recent years, there has been a significant increase in the use of marble and glazed tiles as covering or decorative building materials for home interiors. Marble tiles are commonly used as wall and floor covering, facing materials for buildings, kitchen counter-top, vanity tops and inner and outer decorative materials because of their appearance, attractive colours, polished surface and durability against external conditions and high resistance to wear. Turkey's total probable marble reserves are ~13.9 million tonnes, which corresponds to 40 % of the world's total marble reserves⁽¹¹⁾ and the production capacity of marble in Turkey is 6.5 million m²⁽¹²⁾. Turkey is the eighth largest exporter of marble in the world, and it has been exporting marble to European countries such as Germany, Italy, Spain and the Netherlands etc. Glazed tiles are used only in kitchens, bathrooms and toilets as decorative building materials. The Turkish ceramic industry boasts the fifth largest production of ceramic tiles in the world, the major part of which is glazed and unglazed tiles⁽³⁾. Turkey is the third largest exporter of ceramic in the world with its exports of 60 million m². Ceramic tile production of Turkey constitutes 3.5 % of world production and 11 % of European production.

So far, a few studies related to the radioactivity in covering or decorative materials used in Turkey have been published in the literature^(3, 12, 13). However, the

detailed information of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in decorative building materials in Turkey is not available in the literature.

The goal of the present study firstly was to measure the activity concentrations of primordial radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in 77 different samples of two types covering building materials (marble and glazed tiles) were measured by using gamma-ray spectrometer with high-purity germanium (HPGe). Secondly, the goal was to evaluate the potential radiological hazards caused from the usage of these materials in construction sectors were

evaluated by calculating the external exposure index (activity concentration index), the internal exposure index (alpha index), the indoor absorbed gamma dose rate (AGDR) and the corresponding annual effective dose (AED). The results were presented in tables and figures, discussed on the basis of relevant national and international legislation and guidance and compared with the similar results obtained for marble and glazed tiles in the literature.

MATERIALS AND METHODS

Sample preparation for radioactivity measurements

One sample of each of the various types of marble and glazed tiles (38 marble tiles and 39 glazed tiles) is obtained from major manufacturers and suppliers in Turkey. All samples were crushed and then dried in a temperature-controlled furnace at 105°C for 15–20 h to remove moisture. Each of them was homogenised and then filled into cylindrical plastic containers of 60 mm height and 50 mm diameter. The geometrical dimensions of the samples were kept identical to those of the reference materials. The weighed containers were hermetically sealed and stored for >4 weeks to capture ^{222}Rn gas and ensure secular equilibrium between ^{226}Ra and its decay products as well as between ^{228}Ra and its short-lived decay products.

Radioactivity measurement system and detector calibration

Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the studies' covering building material samples were measured by using a high-resolution gamma-ray spectrometric system⁽¹⁴⁾. The system was consisted of a coaxial p-type HPGe detector shielded from background radiation. The detector was interfaced to the digital spectrum analyzer (DSA-1000), which was a full-featured, 16 K channel multichannel analyzer on advanced digital signal processing techniques. DSA-1000 operates through Genie-2000 gamma spectroscopy software, including peak searching, peak evaluation, energy/efficiency calculation mode, nuclide identification, etc.

Calibration of the detector

The efficiency calibration of the HPGe detector was performed using the radionuclide-specific efficiency method in which the efficiency values of gamma-ray lines belonging to the specific radionuclide existing in both the reference material and sample were only used. Thus, the uncertainty in gamma-ray intensities, the influence of coincidence-summing correction and self-absorption effects of the emitting gamma photons were avoided for the close geometry

Table 1. The values of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K measured in the 38 different marble tile samples.

Sample code	Activity concentration (Bq kg ⁻¹)		
	^{226}Ra	^{232}Th	^{40}K
MRBT1	3.5 ± 0.1	3.0 ± 0.2	56.2 ± 4.8
MRBT2	11.7 ± 0.6	4.3 ± 0.2	74.2 ± 6.2
MRBT3	7.1 ± 0.3	4.2 ± 0.3	60.9 ± 4.9
MRBT4	41.3 ± 1.8	82.0 ± 2.6	729.3 ± 55.6
MRBT5	56.3 ± 2.9	3.1 ± 0.2	21.1 ± 2.0
MRBT6	3.3 ± 0.2	3.2 ± 0.2	16.0 ± 1.6
MRBT7	29.4 ± 1.4	2.4 ± 0.2	34.4 ± 3.1
MRBT8	2.3 ± 0.2	3.1 ± 0.3	13.2 ± 1.1
MRBT9	23.4 ± 0.9	9.4 ± 0.5	98.3 ± 8.4
MRBT10	2.0 ± 0.2	1.4 ± 0.1	13.4 ± 1.2
MRBT11	1.9 ± 0.2	1.2 ± 0.1	14.0 ± 1.2
MRBT12	2.1 ± 0.2	1.3 ± 0.1	12.0 ± 1.1
MRBT13	6.1 ± 0.4	1.0 ± 0.1	21.4 ± 1.9
MRBT14	7.2 ± 0.4	3.1 ± 0.3	13.2 ± 1.2
MRBT15	4.0 ± 0.3	2.4 ± 0.2	16.4 ± 0.4
MRBT16	1.4 ± 0.2	1.3 ± 0.1	20.1 ± 0.3
MRBT17	1.3 ± 0.2	1.4 ± 0.1	18.0 ± 1.5
MRBT18	2.4 ± 0.2	2.1 ± 0.2	29.3 ± 0.5
MRBT19	5.4 ± 0.4	2.3 ± 0.2	15.4 ± 1.3
MRBT20	1.9 ± 0.1	1.8 ± 0.1	18.4 ± 0.4
MRBT21	1.3 ± 0.1	2.2 ± 0.2	18.4 ± 0.4
MRBT22	2.4 ± 0.2	2.3 ± 0.2	31.4 ± 0.3
MRBT23	3.2 ± 0.2	3.4 ± 0.2	29.2 ± 2.5
MRBT24	15.0 ± 0.6	12.3 ± 0.7	367.2 ± 26.8
MRBT25	1.3 ± 0.1	1.4 ± 0.1	16.2 ± 0.5
MRBT26	5.0 ± 0.4	10.2 ± 0.7	18.4 ± 1.6
MRBT27	6.4 ± 0.4	4.4 ± 0.3	16.3 ± 0.6
MRBT28	2.4 ± 0.2	3.4 ± 0.2	13.3 ± 0.5
MRBT29	2.3 ± 0.2	2.4 ± 0.2	22.2 ± 0.3
MRBT30	3.4 ± 0.4	2.4 ± 0.2	20.4 ± 0.4
MRBT31	3.1 ± 0.3	4.4 ± 0.3	20.3 ± 0.4
MRBT32	3.2 ± 0.2	2.3 ± 0.3	18.4 ± 0.4
MRBT33	2.1 ± 0.2	3.2 ± 0.3	18.0 ± 0.3
MRBT34	32.3 ± 1.4	1.4 ± 0.1	20.4 ± 1.8
MRBT35	2.2 ± 0.2	3.4 ± 0.2	191.4 ± 13.8
MRBT36	2.3 ± 0.3	1.4 ± 0.2	26.1 ± 2.0
MRBT37	1.4 ± 0.2	1.3 ± 0.4	42.2 ± 5.9
MRBT38	8.4 ± 1.3	11.3 ± 2.3	22.3 ± 0.7

conditions. The reference materials RGU-1 (U-ore), RGTh-1 (Th-ore) and RGK-1 (K₂SO₄) used for the radionuclide-specific efficiency calibration of the counting system were counted by placing them in the sample position. The efficiencies of gamma-ray line of 295.2, 351.9, 609.3, 583.2, 911.2, 1120.3, 1238.1, 1764.5 and 1460.8 keV were calculated using the following formula:

$$\varepsilon(E_\gamma) = \frac{N}{P_\gamma \cdot t \cdot A} \quad (1)$$

where N is the corrected net peak area of the corresponding gamma-ray photopeak at energy E_γ

emitted by a radionuclide, A the activity of the calibration source or reference material (in Bq), P_γ the emission probability and t live time of the sample spectrum in seconds. Then the measured efficiency values were fitted to function as follows:

$$\varepsilon(E_\gamma) = \frac{1}{E_\gamma} [a + b \cdot \ln(E_\gamma) + c \cdot \ln(E_\gamma)^2 + d \cdot \ln(E_\gamma)^3] \quad (2)$$

where E_γ is gamma-ray energy, and a, b, c and d are the fitted parameters and correspond to $-29.010, 10.944, -0.851$ and -0.004 , respectively.

Table 2. The values of the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K measured in the 39 glazed tile samples.

Sample code	Activity concentration (Bq kg ⁻¹)		
	²²⁶ Ra	²³² Th	⁴⁰ K
GLZT1	92.8 ± 11.0	75.7 ± 5.0	588.7 ± 21.0
GLZT2	85.4 ± 12.0	84.2 ± 6.0	523.4 ± 29.0
GLZT3	195.2 ± 17.0	105.4 ± 5.0	484.9 ± 37.2
GLZT4	137.7 ± 14.0	114.8 ± 4.0	466.8 ± 25.7
GLZT5	57.3 ± 7.6	103.4 ± 10.1	17.0 ± 1.5
GLZT6	111.7 ± 9.0	48.8 ± 3.0	321.9 ± 21.1
GLZT7	106.7 ± 3.0	70.6 ± 1.0	362.5 ± 26.3
GLZT8	64.8 ± 1.0	46.8 ± 1.0	192.6 ± 12.9
GLZT9	76.2 ± 4.0	89.1 ± 2.0	640.2 ± 36.7
GLZT10	93.2 ± 10.0	49.4 ± 4.0	392.4 ± 18.0
GLZT11	53.1 ± 11.0	42.2 ± 4.0	415.4 ± 25.0
GLZT12	70.4 ± 2.8	58.2 ± 3.1	362.4 ± 26.1
GLZT13	80.1 ± 2.3	59.0 ± 2.9	477.9 ± 28.5
GLZT14	65.3 ± 1.6	68.2 ± 2.5	579.0 ± 37.1
GLZT15	40.4 ± 2.4	53.4 ± 4.0	290.1 ± 8.9
GLZT16	96.1 ± 4.3	69.2 ± 5.0	561.0 ± 35.7
GLZT17	116.2 ± 15.6	73.2 ± 5.9	456.3 ± 62.0
GLZT18	50.0 ± 2.6	60.0 ± 3.4	980.0 ± 89.0
GLZT19	104.3 ± 6.8	41.2 ± 3.0	425.2 ± 18.0
GLZT20	84.2 ± 3.5	55.7 ± 4.0	468.6 ± 23.0
GLZT21	73.2 ± 2.3	57.8 ± 3.0	526.7 ± 22.0
GLZT22	37.8 ± 2.4	53.2 ± 2.4	384.0 ± 21.2
GLZT23	40.2 ± 2.1	51.0 ± 1.9	411.3 ± 17.9
GLZT24	80.9 ± 4.4	61.8 ± 2.0	356.7 ± 16.0
GLZT25	38.1 ± 1.9	45.3 ± 3.9	160.0 ± 9.6
GLZT26	45.2 ± 2.9	71.2 ± 6.7	179.0 ± 9.8
GLZT27	171.1 ± 6.1	80.2 ± 8.5	21.3 ± 1.8
GLZT28	61.2 ± 2.1	51.4 ± 2.3	414.2 ± 26.4
GLZT29	75.0 ± 3.1	57.0 ± 3.2	562.3 ± 35.3
GLZT30	68.3 ± 2.0	61.7 ± 4.0	425.1 ± 32.7
GLZT31	83.1 ± 2.5	51.3 ± 4.4	365.1 ± 28.2
GLZT32	91.2 ± 3.1	100.4 ± 7.6	694.0 ± 38.5
GLZT33	54.3 ± 1.3	48.0 ± 2.8	325.7 ± 14.5
GLZT34	61.9 ± 2.8	48.1 ± 2.2	632.4 ± 29.5
GLZT35	84.9 ± 3.5	76.1 ± 4.3	572.3 ± 28.2
GLZT36	81.2 ± 3.8	73.1 ± 2.7	578.4 ± 37.1
GLZT37	84.4 ± 5.1	57.0 ± 3.3	589.0 ± 31.4
GLZT38	105.2 ± 6.7	88.3 ± 4.2	1044.0 ± 57.4
GLZT39	49.4 ± 2.5	48.2 ± 3.4	306.1 ± 13.5

Measurement of activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K

The sample containers were placed on top of the detector for counting. The counting time ranged from 20 000 to 86 400 s. Gamma-ray background at the laboratory was determined to correct the net counts for the sample. The activity concentration (A) (Bq kg⁻¹) of radionuclides mentioned above was calculated from the following equation:

$$A = \frac{N}{\varepsilon(E_\gamma) \cdot P_\gamma \cdot t \cdot M} \quad (3)$$

where N is the net peak area of gamma-ray at energy E_γ subtracted from background, $\varepsilon(E_\gamma)$ is the absolute efficiency for gamma rays, P_γ is the gamma-ray yield per decay, t is the counting live time and M is the dried sample mass in terms of kilogram.

Table 3. The statistical data for the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K (Bq kg⁻¹) measured in the examined tile samples.

Material	Descriptive statistical data (Bq kg ⁻¹)			
	²²⁶ Ra	²³² Th	⁴⁰ K	
Marble	Mean	8.2	5.5	58.1
	Median	3.2	2.4	20.4
	Min	1.3	1.0	12.0
	Max	56.3	82.0	729.3
	Standard deviation	12.3	13.1	128.5
	Standard error	2.0	2.1	20.9
	Kurtosis	6.7	34.4	21.5
Glazed tile	Skewness	2.6	5.7	4.5
	Mean	81.2	65.4	450.1
	Median	80.1	59.0	425.2
	Min	37.8	41.2	17.0
	Max	195.2	114.8	1044.0
	Standard deviation	33.6	18.8	205.6
	Standard error	5.4	3.0	32.9
Kurtosis	3.1	0.3	1.9	
Skewness	1.5	1.0	0.5	

The activity concentrations were averaged from gamma-ray photopeaks at several energies assuming the secular equilibrium between ^{226}Ra and ^{222}Rn and ^{232}Th and ^{228}Ra . The activity concentration of ^{226}Ra was derived from the gamma-rays of 351.9 keV (^{214}Pb) and 609.3 keV (^{214}B), while the activity concentration of ^{232}Th was derived from the gamma-rays of the 911.2 keV (^{228}Ac) and 583.2 keV (^{208}Tl). The activity concentration of ^{40}K was measured directly by its gamma-ray at 1460.8 keV

The minimum detectable activity concentration (MDC) (Bq kg^{-1}) of the gamma-ray measurements was calculated as follows⁽¹⁴⁾:

$$\text{MDC} = \frac{F_C \cdot \sigma_B}{\varepsilon(E_\gamma) \cdot P_\gamma \cdot t \cdot M} \quad (4)$$

where F_C is the statistical coverage factor—equal to 1.64 (confidence level 95%), σ_{N_B} is the standard

deviation of the background in the region of interest and equals square root of the number of counts for the background spectrum, $\varepsilon(E_\gamma)$ is the absolute efficiency of the detector, P_γ is the gamma-ray yield per decay, t is the counting live time in seconds and M is the dried sample mass in terms of kilogram. The mean value of MDC for ^{226}Ra , ^{232}Th and ^{40}K was estimated to be 0.4, 0.5 and 2.4 Bq kg^{-1} for marble tile samples, and 0.6, 0.8 and 2.9 Bq kg^{-1} for glazed tile samples, respectively.

The uncertainty of the activity concentration (ΔA) is calculated by the following formula:

$$\frac{\Delta A}{A} = \sqrt{\frac{\Delta N_R}{N_R} + \frac{\Delta P_\gamma}{P_\gamma} + \frac{\Delta \varepsilon}{\varepsilon} + \frac{\Delta M}{M}} \quad (5)$$

where ΔN_R is the count rate uncertainty, ΔP_γ the emission probability uncertainty found in the

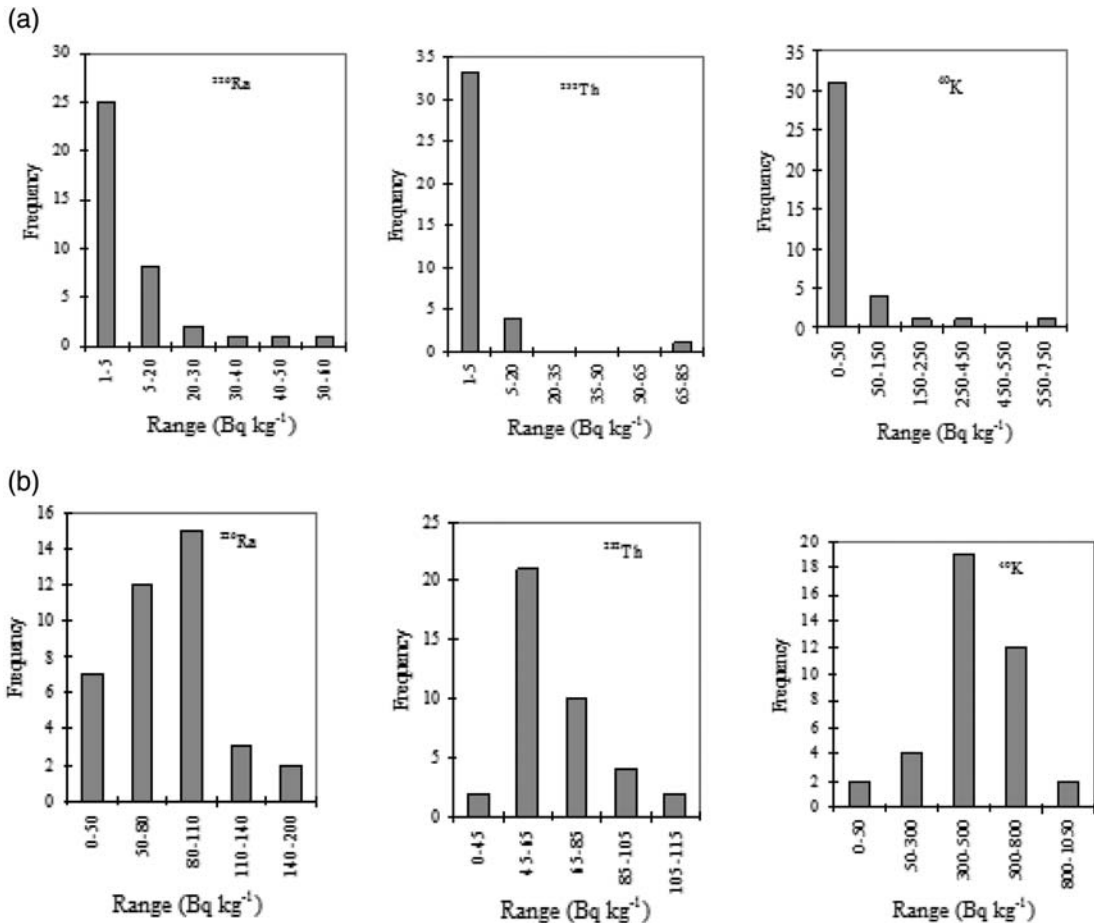


Figure 1. Frequency distribution of the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K for (a) marble and (b) glazed tile samples.

nuclear data tables, $\Delta\epsilon$ the efficiency uncertainty and ΔM the weighing uncertainty.

Dose evaluation

In this study, the external exposure index (activity concentration index), the internal exposure index (alpha index), the indoor AGDR and the corresponding AED were determined to evaluate the radiation dose associated with the usage of the decorative building materials samples.

External exposure index (activity concentration index)

A number of indices related to the evaluation of the external exposure arising from gamma-ray emitting radionuclides in building materials have been proposed by several investigators⁽⁷⁾. In the present study, the activity concentration index (I_γ) proposed by the European Commission (EC) recommendation was taken into consideration and calculated for ready-to-use building materials using the following formula⁽¹⁵⁾:

$$I_\gamma = \frac{A_{Ra}}{300\text{Bq kg}^{-1}} + \frac{A_{Th}}{200\text{Bq kg}^{-1}} + \frac{A_K}{3000\text{Bq kg}^{-1}} \tag{6}$$

where A_{Ra} , A_{Th} and A_K are the activity concentrations

Table 4. Comparison of the mean values of activity concentrations measured in the present study for the samples examined with those obtained in other countries.

Material	Country	Activity concentration (Bq kg ⁻¹)			References
		²²⁶ Ra	²³² Th	⁴⁰ K	
Marble	Nigeria	9.4	5.6	33.4	(17)
	Nigeria	2.4	0.7	6.6	(18)
	Jordan (Ajloun)	19.3	11.9	93.3	(19)
	Serbia	37	19.2	31	(20)
	Pakistan	18	18	299	(21)
	Pakistan	7.9	2.7	25.8	(22)
	Egypt (Quena)	205	115	865	(23)
	Algeria	23	18	310	(24)
	Malaysia	19.0 ^a	16.5	243.3	(25)
	Nigeria	2.4	0.7	6.6	(18)
	Turkey	23	15	149	(26)
	Turkey	8.2	5.5	58.1	This study
	Glazed tile	China	109	102	599
China		91	87	768	(28)
Italy		114.0	55.0	506.0	(29)
Turkey		81.2	65.4	450.1	This study

^aThis value is given for ²³⁸U.

of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively (in Bq kg⁻¹). The activity concentration index should be used only as a screening tool for identifying materials that might be of concern. For superficial and other materials with restricted use (tiles, boards, etc.), $I_\gamma \leq 6$ corresponds to an AED ≤ 1 mSv, which is the dose criterion for controls, while $I_\gamma \leq 2$ corresponds to an AED ≤ 0.3 mSv, which is the dose criterion for exemption.

Internal exposure index (alpha index)

A few indices dealing with the assessment of the excess alpha radiation due to inhalation originating from building materials were developed⁽⁷⁾. In this study, the alpha index (I_α) was calculated by the following formula:

$$I_\alpha = \frac{C_{Ra}}{200\text{Bq kg}^{-1}} \tag{7}$$

where A_{Ra} is the activity concentration of ²²⁶Ra in Bq kg⁻¹. In the Turkish Standard TS 12614, the recommended upper level indoor radon exposure in buildings in Turkey has been given as 200 Bq m⁻³⁽¹⁶⁾. $I_\alpha \leq 1$ corresponds to the ²²⁶Ra activity concentration

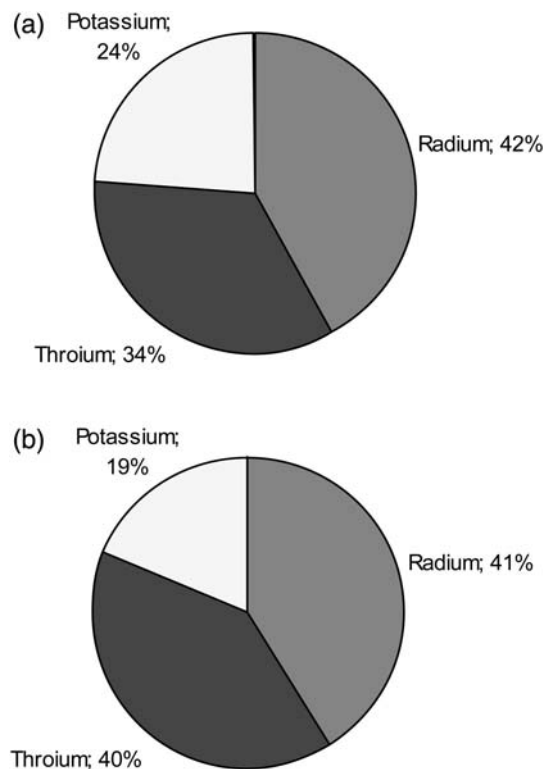


Figure 2. The activity concentration index (a) and the alpha index (b) calculated for marble tile samples.

RADIOACTIVITY LEVELS OF COVERING MATERIALS

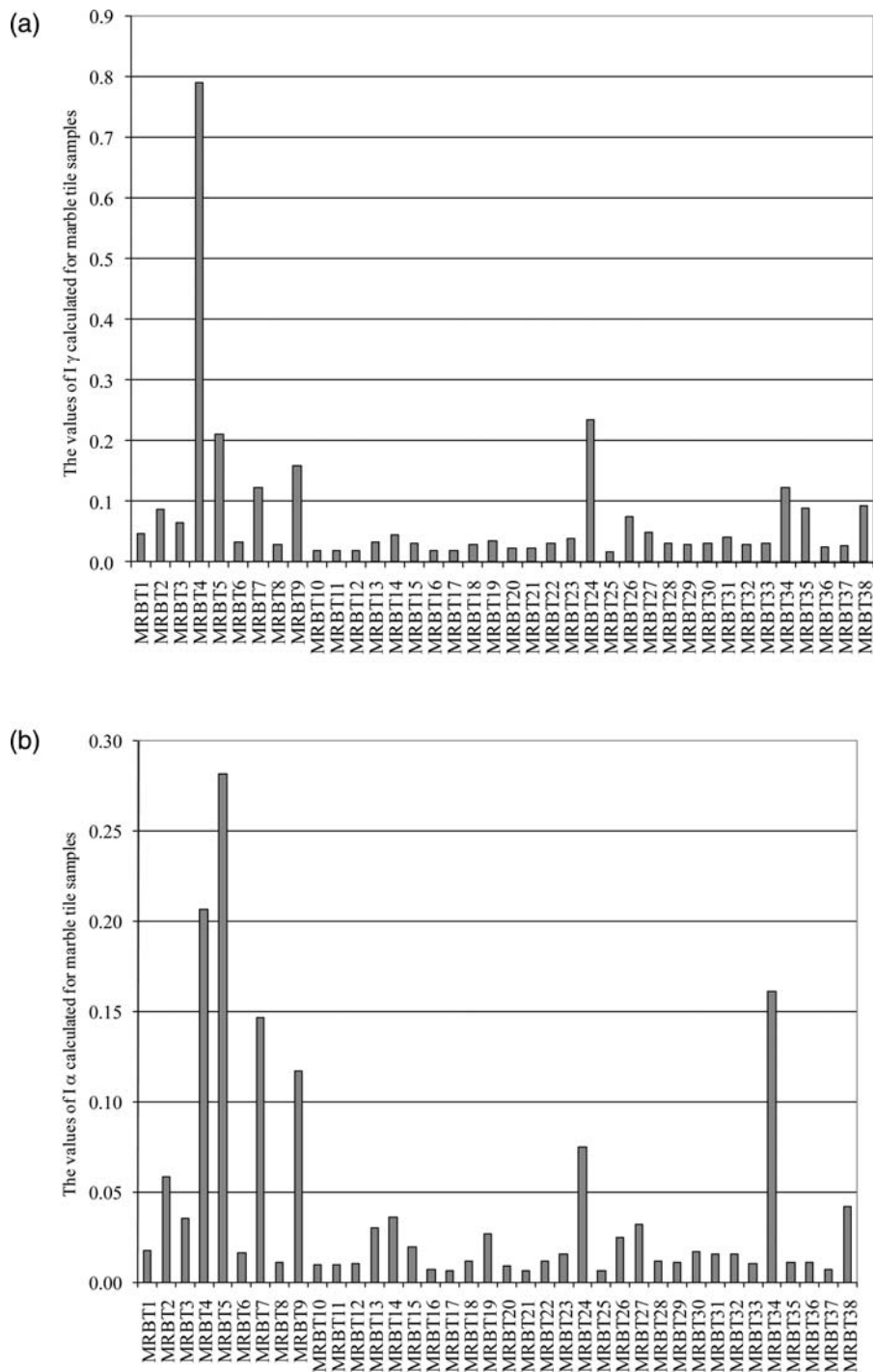


Figure 3. The activity concentration index (a) and the alpha index (b) calculated for glazed tile samples.

$\leq 200 \text{ Bq kg}^{-1}$. When the activity concentration of ^{226}Ra in a building material exceeds the value of 200 Bq kg^{-1} , it is possible that the radon exhalation from this material could cause indoor radon concentration exceeding $200 \text{ Bq m}^{-3(7)}$.

AGDR indoors

It is possible to evaluate the indoor absorbed dose rate due to the external gamma radiation arising from the decorative building samples widely used in construction of dwellings and work places from the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K measured for each sample. In this study, the AGDR (nGy h^{-1}) in indoor air was evaluated using data and formulae provided by the EC report⁽¹⁵⁾. In the EC report, the dose conversion coefficients were calculated for the centre of the standard room. The dimensions of the room are $4 \times 5 \times 2.8 \text{ m}^3$. The thickness of tiles on all walls and density of the structures are 3 cm and 2600 kg m^{-3} , respectively. These coefficients correspond to 0.12 nGy h^{-1} per Bq kg^{-1} for ^{226}Ra , 0.14 nGy h^{-1} per Bq kg^{-1} for ^{232}Th and $0.0096 \text{ nGy h}^{-1}$ per Bq kg^{-1} for ^{40}K :

$$\text{AGDR} = 0.12 \times A_{\text{Ra}} + 0.14 \times A_{\text{Th}} + 0.0096 \times A_{\text{K}} \tag{8}$$

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively in Bq kg^{-1} .

Annual effective dose

The corresponding AED (mSv) indoors was calculated using the following formula:

$$\text{AED} = \text{AGDR} \times \text{OT} \times \text{CF} \times 10^{-6} \tag{9}$$

where AGDR is the absorbed gamma dose rate indoors given by Equation (8), OT is the indoor occupancy time ($0.8 \times 24 \text{ h d}^{-1} \times 365.25 \text{ d y}^{-1}$) and CF is the conversion factor (0.7 Sv Gy^{-1}) from absorbed dose in air to effective dose received by adults. The indoor occupancy factor is 0.8, implying 80 % of time is spent indoors.

RESULTS AND DISCUSSION

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K measured in the examined covering building material samples are shown in Table 1 (marble tiles) and Table 2 (glazed tiles). The descriptive statistical data for the activity concentrations of the radionuclides are shown in Table 3. As can be seen from Tables 1 to 3, the average concentration values of ^{226}Ra , ^{232}Th and ^{40}K measured in the marble tile samples

is significantly lower than the earth's crust averages (population weighted) of 32, 45 and 412 Bq kg^{-1} , respectively⁽¹⁾, while the average concentration values of ^{226}Ra , ^{232}Th and ^{40}K measured in the glazed tile samples are higher than the quoted values. Figure 1 shows the frequency distribution of the radionuclides' concentrations in the samples. The range of $1\text{--}20 \text{ Bq kg}^{-1}$ measured in marble tile samples for ^{226}Ra and ^{232}Th includes most of the samples (87 % for ^{226}Ra and 95 % for ^{232}Th), while the range of $30\text{--}110 \text{ Bq kg}^{-1}$ measured in glazed tile samples for ^{226}Ra and ^{232}Th includes most of the samples (87 % for ^{226}Ra and 95 % for ^{232}Th).

Table 5. The values of the indoor AGDR and the corresponding AED evaluated for the samples of the marble and glazed tile samples.

Sample code	AGDR (nGy h^{-1})	AED (mSv)	Sample code	AGDR (nGy h^{-1})	AED (mSv)
MRBT1	1.4	0.007	GLZT1	27.4	0.13
MRBT2	2.7	0.013	GLZT2	27.1	0.13
MRBT3	2.0	0.010	GLZT3	42.8	0.21
MRBT4	23.4	0.115	GLZT4	37.1	0.18
MRBT5	7.4	0.036	GLZT5	21.5	0.11
MRBT6	1.0	0.005	GLZT6	23.3	0.11
MRBT7	4.2	0.021	GLZT7	26.2	0.13
MRBT8	0.8	0.004	GLZT8	16.2	0.08
MRBT9	5.1	0.025	GLZT9	27.8	0.14
MRBT10	0.6	0.003	GLZT10	21.9	0.11
MRBT11	0.5	0.003	GLZT11	16.3	0.08
MRBT12	0.5	0.003	GLZT12	20.1	0.10
MRBT13	1.1	0.005	GLZT13	22.5	0.11
MRBT14	1.4	0.007	GLZT14	22.9	0.11
MRBT15	1.0	0.005	GLZT15	15.1	0.07
MRBT16	0.5	0.003	GLZT16	26.6	0.13
MRBT17	0.5	0.003	GLZT17	28.6	0.14
MRBT18	0.9	0.004	GLZT18	23.8	0.12
MRBT19	1.1	0.005	GLZT19	22.4	0.11
MRBT20	0.7	0.003	GLZT20	22.4	0.11
MRBT21	0.6	0.003	GLZT21	21.9	0.11
MRBT22	0.9	0.004	GLZT22	15.7	0.08
MRBT23	1.1	0.006	GLZT23	15.9	0.08
MRBT24	7.0	0.035	GLZT24	21.8	0.11
MRBT25	0.5	0.002	GLZT25	12.5	0.06
MRBT26	2.2	0.011	GLZT26	17.1	0.08
MRBT27	1.5	0.008	GLZT27	32.0	0.16
MRBT28	0.9	0.004	GLZT28	18.5	0.09
MRBT29	0.8	0.004	GLZT29	22.4	0.11
MRBT30	0.9	0.005	GLZT30	20.9	0.10
MRBT31	1.2	0.006	GLZT31	20.7	0.10
MRBT32	0.9	0.004	GLZT32	31.7	0.16
MRBT33	0.9	0.004	GLZT33	16.4	0.08
MRBT34	4.3	0.021	GLZT34	20.2	0.10
MRBT35	2.6	0.013	GLZT35	26.3	0.13
MRBT36	0.7	0.004	GLZT36	25.5	0.13
MRBT37	0.8	0.004	GLZT37	23.8	0.12
MRBT38	2.8	0.014	GLZT38	35.0	0.17
			GLZT39	15.6	0.08

RADIOACTIVITY LEVELS OF COVERING MATERIALS

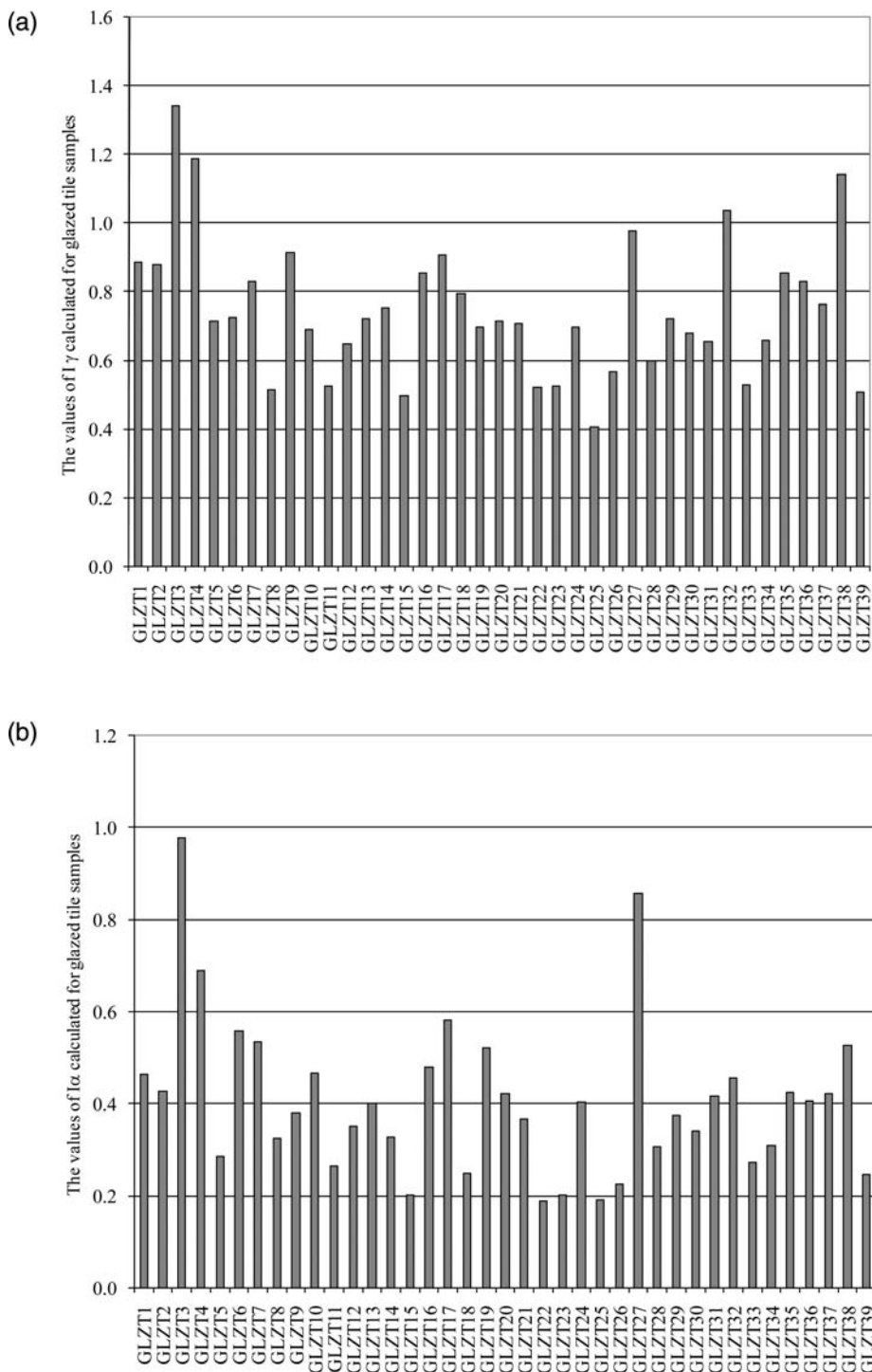


Figure 4. Relative contribution to total indoor AGDR due to ^{226}Ra and ^{232}Th decay products and ^{40}K for (a) marble and (b) glazed tile samples.

The activity concentrations of ^{40}K measured in 90 % of the marble tile samples are between 10 and 150 Bq kg^{-1} , while the activity concentrations of ^{40}K measured in 95 % of the marble tile samples are between 10 and 800 Bq kg^{-1} . Table 4 presents the comparison of the mean values of activity concentrations measured in this study for the samples examined with those obtained in other countries. As can be seen in Table 4, the mean values of the activity concentration of the radionuclides obtained for the marble tile samples are comparable to those obtained from other studies conducted in countries other than Jordan, Egypt, Pakistan and Malaysia, while the mean values of the activity concentration of the radionuclides obtained for the glazed tile sample are comparable to those obtained from other studies.

The average values of the activity concentration index for marble and glazed tile samples are calculated as 0.07 ± 0.02 and 0.75 ± 0.12 , respectively. All values of I_γ calculated for marble and glazed tile samples are shown in Figures 2a and 3a, respectively. It is observed that all values of I_γ for the samples are significantly below the value of 6, corresponding to a dose criterion of 1 mSv. Furthermore, the values of I_γ are lower than the value of 2, corresponding to dose criterion of 0.3 mSv.

The average values of the alpha index for marble and glazed tile samples are calculated as 0.04 ± 0.01 and 0.41 ± 0.07 , respectively. All values of I_α calculated for marble and glazed tile samples are shown in Figures 2b and 3b, respectively. It can be seen that all values of I_α are below the recommended upper level of unity.

The results of the indoor AGDR evaluated for marble and glazed tile samples are given in the second and fifth column of Table 5. From Table 5, it can be seen that all estimated values of AGDR are significantly lower than the world average (populated-weighted) indoor AGDR of $84 \text{ nGy h}^{-1(1)}$. The relative contribution to total absorbed indoor gamma dose rate due to ^{226}Ra and ^{232}Th decay products and ^{40}K for marble and glazed tile samples is shown in Figure 4. The contributions of ^{226}Ra , ^{232}Th and ^{40}K to the total indoor AGDR are 42, 34 and 24 % for marble tile samples and 41, 40 and 19 % for glazed tile samples.

The values of the corresponding AED evaluated for marble and glazed tile samples are given in the third and sixth column of Table 5. From Table 5, it can be seen that all the values of AED are lower than the exemption dose criterion of 0.3 mSv.

CONCLUSION

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in examined marble and glazed tile samples utilised as covering or decorative building materials in

Turkey are determined using high-resolution gamma-ray spectroscopy. The average activity concentration values of ^{226}Ra , ^{232}Th and ^{40}K measured in the marble tile samples is significantly lower than the earth's crust averages, while the average concentration values of ^{226}Ra , ^{232}Th and ^{40}K measured in the glazed tile samples are higher than the quoted values. The measured average concentration values of ^{226}Ra , ^{232}Th and ^{40}K are comparable with the values in other countries. The activity concentration index, the alpha index, the indoor AGDR and the annual indoor effective dose (which is estimated only for comparison purposes), based on models from European Commission Report 112, are also calculated to evaluate the associated potential radiological hazards. The annual indoor effective dose rates received by an individual from the examined samples are significantly lower than the world upper limit value of 1 mSv y^{-1} . Hence, it can be inferred that the utilisation of marble and glazed tile samples as covering or decorative building materials is unlikely to pose any significant adverse health impacts, based on the cited national and international legislation and guidance used for comparison.

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