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Natural radioactivity in ceramic tiles and associated radiological hazards

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ABSTRACT

Background: The natural radioactivity levels in ceramic tiles produced in Turkey were determined and the related radiological hazards were assessed. ***Materials and Methods:*** The natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) in the ceramic tiles were measured using high-purity germanium detector system (HPGe) and the related radiological hazards were assessed by radium equivalent activity (Raeq), external hazard index (Hex), representative level index (Iγ), absorbed dose rate (D) and annual effective dose (AED). ***Results:*** The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the ceramic tiles were found to be 36.59 ± 2.64, 51.23 ± 2.81 and 420.81 ± 12.87 Bq/kg, respectively. These values are less than the world average values except the activity values of 232Th. Again, the averages of radiological hazard parameters (Raeq, Hex, D and AED) found for all ceramic tiles were found lower than the world averages. ***Conclusion:*** The present results was compared with the literature and the international reference values. This study shows that the investigated ceramic tiles can be used safely in constructions and do not create significant radiological hazard when used in constructions.

Keywords: *Ceramic tile, natural radioactivity, radiological hazard, Turkey.*

INTRODUCTION

Construction materials contain natural radioisotopes such as ^{40}K, ^{238}U and ^{232}Th series at various concentrations. People continuously expose to indoor gamma radiations emitting from these radionuclides during their life as long as they stay in closed areas [(1)]. Radium is mostly used as reference instead of uranium since decay series segment starting from radium in the uranium series is radiologically very important [(1)]. Because people spend 80 % of their time indoors (office, home, etc.), it is crucial to know natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) in construction materials in order to determine radiation exposed [(2)]. Therefore, the developed countries support this kind of studies to protect community health and to keep the activity caused by construction materials under a certain level [(3-5)].

The ceramics industry is one of the oldest and fastest growing sectors in Turkey. The sector improves its products and increases the product range every passing day. Turkish ceramics industry is among the significant industries in Turkey with approximately 2 billion US dollar trading volume and nearly 1 billion US dollar export [(6)]. Ceramic tiles are the leading product group of the sector. Today, Turkish ceramic coating materials sector has succeeded in being an expert on tiles manufacturing in the world with the investment made especially after 1990. Turkey ranks as eighth in the manufacturing of ceramic coating materials and ranks as fourth in the export of these materials in the world [(6)]. Ceramic tiles are among decorative construction materials used in the construction of houses and buildings. They are produced by mixing various materials together, cooking the mixture at high temperature, and shaping it.

The present study aims to determine natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) in commercial ceramic tile products manufactured in different regions of Turkey and to investigate potential radiological hazards related to these ceramic tiles.

MATERIALS AND METHODS

Sample collected cities

Ceramic tiles are produced in ten provinces in Turkey. Facilities producing ceramic tiles are located in Eskişehir-Bilecik-Kütahya, İzmir-Manisa-Uşak-Aydın, Çanakkale and Çankırı-Yozgat regions with ratios of 49.82%, 28.61%, 14.61%, 6.96%, respectively [6]. These regions and the number of samples taken from these regions are shown in figure 1. 51 different ceramic tile samples as end-user product are taken from these regions. All of the taken samples have the qualities to be used as floor and wall ceramic.

Preparations of samples

Chemical treatment was not applied to any of the ceramic tiles. All samples were crushed by grinding mill (Zhonghe, ZHM-1T) for triturate. The samples were then sieved to obtain a homogenized particle size and weighed. Finally, the samples were filled into uncontaminated empty cylindrical plastic containers of uniform size and these plastic containers were stored 4 weeks before the analysis at air tight condition in order to obtain secular equilibrium between radium and its short lived decay products.

Determination of radioactivity

Natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) in the ceramic tiles were determined using high-purity germanium detector system (ORTEC HPGe, Model No: GEM55P4-95) with 55% relative efficiency. The resolution (FWHM) of this detector system was 1.9 keV for 1332 keV gamma ray of ^{60}Co. The detector was shielded by a cylindrical lead shield, which had thickness of 10 cm to reduce the background level of the system. Efficiency of the detector was determined with a ^{152}Eu source of known activity. ^{152}Eu source has been widely used for calibration and efficiency determination due to their large range of energies (122, 244, 344, 411, 443, 779, 964, 1112 and 1408 keV) with emission probabilities of 3–29% [7]. An ideal measuring geometry of cylindrical source (homogeneously distributed activity with constant volume and distance) was placed coaxially with the detector for the efficiency determination and the same procedure was applied for the sample measurements.

The activity concentration of ^{226}Ra were determined by averaging of the measured concentrations for ^{214}Pb (295 and 351 keV gamma-ray energies) and ^{214}Bi (609 and 1120 keV gamma-ray energies). The activity concentration of ^{232}Th were determined by averaging of the measured concentrations for ^{212}Pb (238 keV gamma-ray energy), ^{228}Ac (338 and 911 keV gamma-ray energies) and ^{208}Tl (583 keV gamma-ray energy) [8]. The activity concentration of ^{40}K was determined directly from its gamma-ray energy of 1460 keV.

Each sample was placed on the top of the detector and counted for 50000 s. The gamma spectra were analyzed using Gamma Vision which is a data acquisition and analysis program.

To remove the contribution of background from measurements, an empty plastic container was counted in the same measurement condition as the samples.

After measurements and subtraction of the background, the activity concentrations of the radionuclides were calculated using equation 1.

$$A(Bq/kg) = \frac{C_s}{\varepsilon . P_\gamma . m . t} \quad (1)$$

where C_s is the count rate under the corresponding peak, Ɛ is the detector efficiency at the corresponding peak energy, P_γ is the absolute transition probability of the specific γ-ray at the corresponding peak energy, m is the mass of the sample (kg) and t is the counting time (s).

Assessment of the radiological hazards

To compare the radiological effects of ceramic tiles containing different amounts of

^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent activity (Ra_{eq}) value is used and this value is a weighed sum of activities of ^{226}Ra, ^{232}Th and ^{40}K based on the estimation that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4180 Bq/kg of ^{40}K produce the same gamma-ray dose rate. Ra_{eq} is calculated using Equation 2 [9].

$$Ra_{eq}=A_{Ra}+1.43A_{Th}+0.077A_{K} \quad (2)$$

where A_{Ra}, A_{Th} and A_{K} are activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

External radiation exposure due to ^{226}Ra, ^{232}Th and ^{40}K is defined as the external hazard index (H_{ex}) and calculated by equation 3 using the model proposed by Krieger [10].

$$H_{ex}=A_{Ra}/370+A_{Th}/259+A_{K}/4810 \quad (3)$$

where A_{Ra}, A_{Th} and A_{K} are activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Not to create any radiological risk for population of the ceramic products, the values of H_{ex} should be less than unity.

Representative level index (Iγ), used to assess the level of gamma radiation hazards related with the natural radionuclides is calculated by the equation 4 [9].

$$I\gamma=A_{Ra}/300+A_{Th}/200+A_{K}/3000 \quad (4)$$

where A_{Ra}, A_{Th} and A_{K} are activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Besides, the values 300, 200 and 3000 are factors of a dose criterion of 1 mSv/y given by the European Commission Report [11] for ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq/kg. Iγ value is correlated with the annual effective dose rate due to excess external gamma radiation caused by building materials. The Iγ value calculated for the samples must be less or equal to 6. Otherwise, the samples have dose rate higher than 1 mSv/y which is the upper limit of annual effective dose rate for population.

The absorbed dose rate (D) in nGy/h, in air at 1 m above of floor because of natural activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K was calculated using the equation 5 [12].

$$D=0.427A_{Ra}+0.662A_{Th}+0.0432A_{K} \quad (5)$$

Where A_{Ra}, A_{Th} and A_{K} are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively.

The annual effective dose (AED) is calculated using the equation 6 to determine the health effect of the absorbed dose rates.

$$AED(mSv/y)=D\times DCF\times OF\times T \quad (6)$$

Where D is absorbed dose rate in air (nGy/h), DCF is dose conversion factor (0.7 Sv/Gy), OF is indoor occupancy factor (0.8) and T is annual exposure time (8760 h/y) [12].

Figure 1. The provinces where ceramic samples are taken in Turkey (with company and sample numbers).

RESULTS AND DISCUSSION

Specific activities

The minimum detectable activity of the system for ^{226}Ra, ^{232}Th and ^{40}K radioisotopes was calculated as 0.42, 0.48 and 3.88 Bq/kg, respectively. All the activity values calculated for the ceramic tiles are above MDA. Natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) of fifty-one different ceramic tiles samples produced by eleven different company in six province of Turkey have been measured and the found results are given in Table 1 for the ceramic tiles manufactured by companies and table 2 for the produced ceramic tiles in provinces. The natural radioactivity levels found for these 51 ceramic tiles samples are also shown in figure 2. The activity concentrations found in the this study ranged from 18.32 ± 1.82 (produced ceramic tile sample by C4 company in Bilecik province) to 64.50 ± 3.10 Bq/kg (produced ceramic tile sample by C5 company in Çanakkale province) for ^{226}Ra, from 17.38 ± 1.85 (produced ceramic tile sample by C10 company in Uşak province) to 103.76 ± 3.51 Bq/kg (produced ceramic tile sample by C5 company in Çanakkale province) for ^{232}Th and from 90.28 ± 5.79 (produced ceramic tile sample by C5 company in Çanakkale province) to 794.75 ± 17.35 Bq/kg (produced ceramic tile sample by C5 company in Çanakkale province) for ^{40}K. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in investigated 51 ceramic tiles samples were found as 36.59 ± 2.64, 51.23 ± 2.81 and 420.81 ± 12.87 Bq/kg. The world average specific activity values of ^{226}Ra, ^{232}Th and ^{40}K in the building materials are 50, 50, 500 Bq/kg, respectively. The obtained activity values in this study are less than the world average values except the activity values of ^{232}Th. The concentrations of natural radioactivity in the examined ceramic tiles were found to be close to each other in the neighboring regions but at different values in the remote regions (figure 1). This situation proves the natural radioactivity concentrations in the ceramic tiles vary depending on region where raw material of ceramic tiles is mined.

A comparison with this study and other studies conducted in different regions of the world is given in table 3. As shown in Table 3, the found average activity value (36.59 Bq/kg) for ^{226}Ra in this study is lower than reported values in China (73 Bq/kg), Egypt (126 Bq/kg), Yemen (131.88 Bq/kg), Italy (114 Bq/kg), Turkey (77.8 Bq/kg) and Iran (62 Bq/kg) but higher than reported values in India (28.2 Bq/kg) and Cameron (12 Bq/kg). Again, the found average activity value (51.23 Bq/kg) for ^{232}Th in this study is only higher than the reported values in Cameron (20 Bq/kg), but lower than the others. Besides, the found average activity value for ^{40}K is lower than reported values in China (480 Bq/kg), Italy (506 Bq/kg), Turkey (485.9 Bq/kg) and Iran (810 Bq/kg) but higher than reported values in India (24.3 Bq/kg), Egypt (300 Bq/kg), Yemen (400.7 Bq/kg) and Cameron (319 Bq/kg).

Assessment of radiological hazards

To assess the radiological hazards originating from natural radionuclides of examined ceramic tiles in this study, radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), representative level index (I_γ), absorbed dose rate (D) and annual effective dose (AED) values were calculated. The calculated results of Ra_{eq}, H_{ex}, I_γ, D and AED are presented in Table 4 for the ceramic tiles manufactured by companies and Table 5 for the produced ceramic tiles in provinces. Besides, the radiological hazard values (Ra_{eq}, H_{ex}, I_γ, D and AED) calculated for all of the ceramic samples are shown in Figure 3. The calculated Ra_{eq} values for the studied ceramic tiles range from 63.31 Bq/kg to 264.75 Bq/kg and all of these values are lower than the recommended limit of 370 Bq/kg for building materials [12].

As seen in table 4 and Table 5, the maximum (0.72) and minimum (0.17) values of H_{ex} were found in the ceramic tile samples belonging to C5 (Çanakkale) and C10 (Uşak) companies. All values found of H_{ex} values for the ceramic tiles are lower than unity. Hence, it can be concluded that these ceramic tiles are safe to use in the buildings.

The I_γ values ranged from 0.23 to 0.96 and average value was found to be 0.52 for the studied ceramic tile. Table 4 and Table 5 indicate that the $I_γ$ values are less than 6. Therefore, all of the ceramic tiles can be used as building materials without creating any significant radiological threat to the public.

The estimated absorbed dose rate (D) values for the investigated ceramic tiles samples range from 27.70 to 125.34 nGy/h. The D value of only 12 in the studied ceramic tiles (all of the 51 samples) is higher than the world average indoor absorbed gamma dose rate of 84 nGy/h [12] but the average value of D for all of the samples was 67.72 nGy/h and this value is lower than the world average value.

The obtained values for the annual effective dose (AED) are presented in column 6 of table 4 and table 5. As shown in the tables, annual effective dose values vary from 0.15 to 0.61 mSv/y with an average value of 0.33 mSv/y. This average value is lower than the 1 mSv/y limit set by the European Commission [11]. The calculated annual effective dose values showed that ceramic tiles used in this study and produced in Turkey could be used in buildings without any radiological risk to the public.

Table 1. Natural radioactivity levels in the ceramic tiles manufactured by the companies

Company Code	N	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
C1	2	33.08 ± 1.78	34.18 ± 1.96	244.42 ± 10.03
C2	2	29.85 ± 1.74	29.80 ± 1.81	251.61 ± 9.48
C3	7	30.18 ± 2.44	38.36 ± 2.44	343.98 ± 11.66
C4	7	28.70 ± 2.53	55.34 ± 2.93	410.09 ± 13.28
C5	10	49.36 ± 3.04	68.40 ± 3.22	501.51 ± 13.40
C6	2	30.09 ± 1.94	47.33 ± 2.27	448.99 ± 13.21
C7	5	32.16 ± 2.86	38.72 ± 2.88	422.77 ± 13.97
C8	4	38.88 ± 3.21	65.71 ± 3.51	586.01 ± 16.55
C9	5	33.88 ± 2.50	50.11 ± 2.74	448.28 ± 13.74
C10	4	39.09 ± 2.62	52.94 ± 2.80	316.71 ± 10.65
C11	3	44.11 ± 2.78	43.87 ± 2.72	437.18 ± 11.65
Average*	51	36.59 ± 2.64	51.23 ± 2.81	420.81 ± 12.87
Minimum*	51	18.32	17.38	90.28
Maximum*	51	64.50	103.76	794.75

* For the total samples

Table 2. Natural radioactivity levels in the ceramic tiles produced in the provinces

Province	N	Statistics	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Bilecik	18	Average	29.89 ± 2.32	43.55 ± 2.51	348.36 ± 11.86
		Min - Max	18.32 - 40.32	23.72 - 63.85	163.79 - 506.39
Çanakkale	10	Average	49.36 ± 3.04	68.40 ± 3.22	501.51 ± 13.40
		Min - Max	26.65 - 64.50	23.81 - 103.76	90.28 - 794.75
Eskişehir	7	Average	31.57 ± 2.60	41.18 ± 2.70	430.26 ± 13.76
		Min - Max	24.95 - 36.97	31.69 - 57.04	180.35 - 523.62
İzmir	4	Average	38.88 ± 3.21	65.71 ± 3.51	586.01 ± 16.55
		Min - Max	35.17 - 43.63	56.72 - 75.72	454.10 - 705.34
Kütahya	5	Average	33.88 ± 2.50	50.11 ± 2.74	448.28 ± 13.74
		Min - Max	25.61 - 44.37	30.69 - 67.36	359.96 - 555.59
Uşak	7	Average	41.24 ± 2.69	49.05 ± 2.77	368.34 ± 11.08
		Min - Max	21.21 - 51.24	17.38 - 73.48	172.07 - 663.60

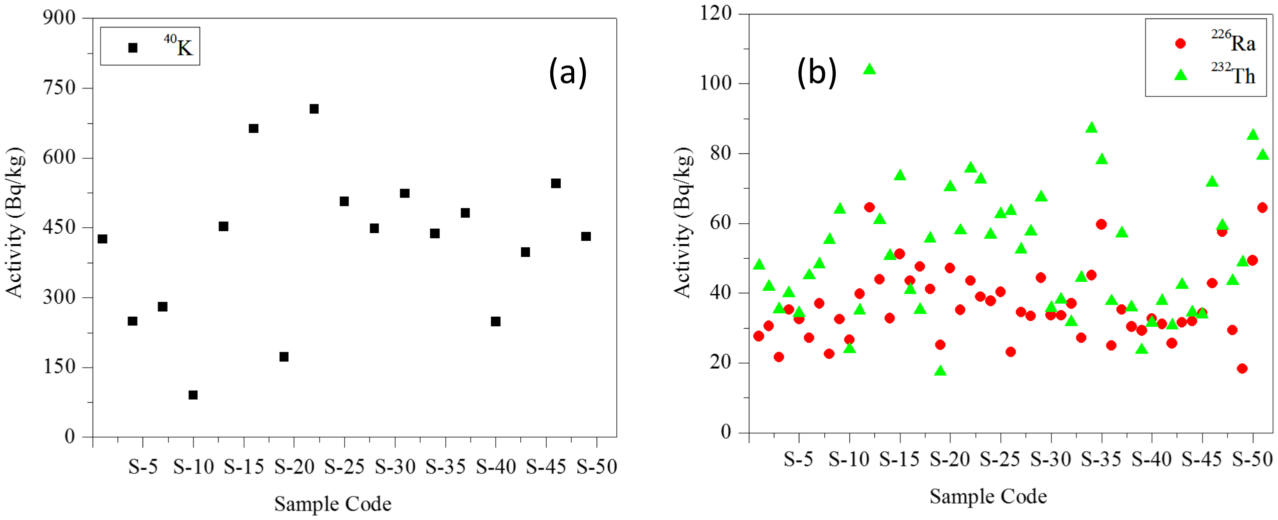

Figure 2. ^{40}K **(a)**, ^{226}Ra and ^{232}Th **(b)** concentrations of the ceramic tile samples.

Table 3. The average natural activity concentrations in ceramic tile samples in Turkey and some other countries.

Country of Origin	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Turkey (Present study)	36.59	51.23	420.81
India [13]	28.2	63.7	24.3
China [14]	73	62	480
Egypt [15]	126	72	300
Yemen [16]	131.88	83.55	400.7
Cameron [17]	12	20	319
Italy [18]	114	55	506
Turkey [19]	77.8	62.9	485.9
Iran [20]	62	54	810

Table 4. The calculated values of radiological hazard parameters in the ceramic tiles manufactured by the companies

Company Code	Ra_{eq} (Bq/kg)	H_{ex}	I_g	D (nGy/h)	AED (mSv/y)
C1	100.77	0.27	0.36	87.58	0.43
C2	91.84	0.25	0.33	80.37	0.39
C3	111.52	0.30	0.41	97.48	0.48
C4	139.41	0.38	0.51	120.08	0.59
C5	185.79	0.50	0.67	160.77	0.79
C6	132.35	0.36	0.49	115.67	0.57
C7	120.09	0.32	0.44	106.00	0.52
C8	177.97	0.48	0.65	154.93	0.76
C9	140.06	0.38	0.51	122.15	0.60
C10	139.18	0.38	0.50	119.53	0.59
C11	140.50	0.38	0.51	123.81	0.61
Average*	142.26	0.38	0.52	67.72	0.33
Minimum*	63.31	0.17	0.23	29.70	0.15
Maximum*	264.75	0.72	0.96	125.34	0.61

*For all the ceramic tile samples

Table 5. The calculated values of radiological hazard parameters in the ceramic tiles produced in the provinces

Province	Statistics	Ra_{eq} (Bq/kg)	H_{ex}	I_g	D (nGy/h)	AED (mSv/y)
Bilecik	Average	118.85	0.32	0.43	56.51	0.28
	Minimum	75.81	0.20	0.27	32.28	0.17
	Maximum	168.79	0.46	0.62	80.52	0.39
Çanakkale	Average	185.79	0.50	0.67	88.02	0.43
	Minimum	67.64	0.18	0.24	31.04	0.15
	Maximum	264.75	0.71	0.96	125.34	0.61
Eskişehir	Average	123.59	0.33	0.46	59.33	0.29
	Minimum	96.18	0.26	0.34	44.56	0.22
	Maximum	153.88	0.42	0.56	73.61	0.36
İzmir	Average	177.97	0.48	0.65	85.42	0.42
	Minimum	152.92	0.41	0.56	72.96	0.36
	Maximum	206.22	0.56	0.76	99.23	0.49
Kütahya	Average	140.06	0.38	0.51	67.01	0.33
	Minimum	97.21	0.26	0.36	46.80	0.23
	Maximum	183.48	0.50	0.67	87.54	0.43
Uşak	Average	139.75	0.38	0.51	66.00	0.32
	Minimum	63.31	0.17	0.23	29.70	0.15
	Maximum	187.65	0.51	0.67	88.10	0.43

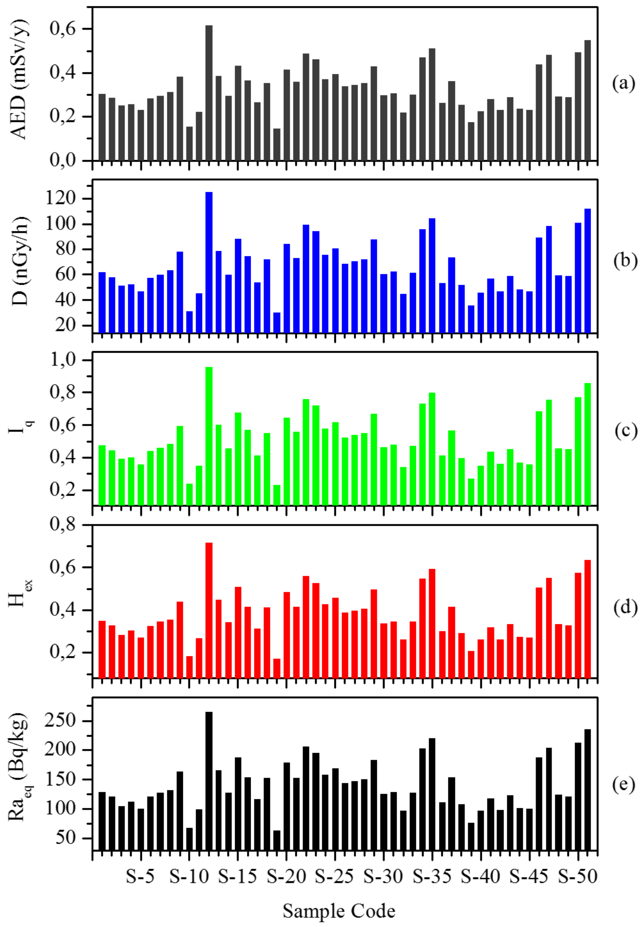

Figure 3. Annual effective dose **(a)**, absorbed dose rate **(b)**, representative level index **(c)**, external hazard index **(d)** and radium equivalent activity **(e)** values calculated for all of the ceramic tile samples.

CONCLUSION

The natural radioactivity levels (^{226}Ra, ^{232}Th and ^{40}K) and related radiological hazards in commercial ceramic tiles produced in Turkey were determined by gamma-ray spectrometer. In this study, 51 samples of ceramic tiles belonging to 11 companies from 6 provinces were taken and studied. For each of the taken ceramic tiles, specific activity, radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), representative level index (I_γ), absorbed dose rate (D) and annual effective dose (AED) values were calculated.

The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in investigated 51 ceramic tiles samples in this study were found as 36.59 ± 2.64, 51.23 ± 2.81 and 420.81 ± 12.87 Bq/kg. These values are less than the world average values (50, 50, 500 Bq/kg) except the activity values of ^{232}Th. The results of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations of all ceramic tiles samples show that there are considerable variations in natural radionuclide concentrations of the ceramic tiles originating from different region.

As a result, this study show that the investigated ceramic tiles can be used safely in constructions and do not create significant radiological hazard when used in constructions. Also, this study can be used as reference for future studies.

Conflicts of interest: *Declared none.*

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