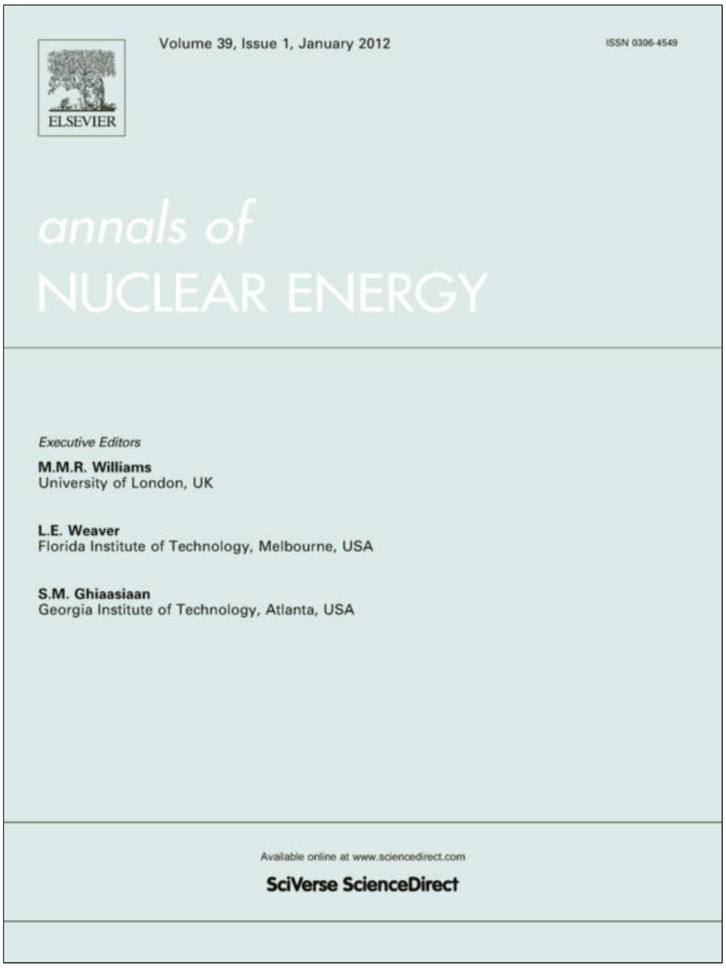

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Estimation of possible radiological hazards from natural radioactivity in commercially-utilized ornamental and countertops granite tiles

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ABSTRACT

The aim of this study is to estimate natural radioactivity levels of commercially-utilized ornamental or countertops granite tiles collected from major retailers in Turkey and possible radiological hazards from usage of these materials by calculating external exposure and internal index, indoor absorbed gamma dose rate and the corresponding annual effective dose. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were determined for 42 kinds of different granite tiles using high-resolution gamma-ray spectrometer. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K varied from 9.2 to 192.5 Bq kg^{-1} (with a mean of 88.4 ± 6.9 Bq kg^{-1}), 7.5 to 344.6 Bq kg^{-1} (with a mean of 95.3 ± 10.0 Bq kg^{-1}) and 92.1 to 4155.9 Bq kg^{-1} (with a mean of 1055.2 ± 103.0 Bq kg^{-1}), respectively. The estimated radiological hazard indices were revised in the light of the relevant national and international legislation and guidance. The values of the radiological hazard indices were found to be within relevant all limit values for superficial materials. The indoor absorbed gamma dose rates estimated from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and the corresponding annual effective doses varied from 4.74 to 85.27 nGy h^{-1} and 0.02 to 0.42 mSv, respectively.

1. Introduction

Granites are igneous rocks which were formed by slowly cooling of magma. Distinct types of granites have different geological origins and mineralogical compositions (Anjos et al., 2005). Granites are mainly composed of feldspar (potassium and sodium) of various colors, coarse grains of quartz and some other minerals (Tzortzis et al., 2003). Granite is hard and tough, but can be shaped and polished, and therefore it has been utilized as building and ornamental materials for interior and exterior. Recently, the granites utilized for kitchen counter and vanity tops and flooring tiles for home interiors are very popular in Turkey.

Granitic rocks contain different amounts of naturally occurring radionuclides of terrestrial origin, ^{238}U and ^{232}Th series and ^{40}K. External and internal exposures indoors and outdoors result from terrestrial radionuclides existing in granite tiles. Thereby the utilization of granite tiles as ornamental and countertops in a home can cause an increase in long-term whole-body exposure of the occupants to ionization radiation emitted from terrestrial radionuclides (Llope, 2011). For this reason, the estimation of the radioactivity levels of granite tiles utilized in construction sectors is crucial in the assessment of possible radiological hazards to human health.

In recent years, there has been a significant increase in the utilization of granites tiles as kitchen counter and vanity tops and flooring tiles for home interiors. Hence, remarkable efforts have been made to determine the radioactivity in granite tiles utilized in the different countries (Abd El-mageed et al., 2011; Marocchi et al., 2011; Anjos et al., 2011; Chen et al., 2010; Myatt et al., 2010; Kitto et al., 2009; El-Arabi et al., 2008; Sakoda et al., 2008; Asghar et al., 2008; Papaefthymiou, 2008; Al-Saleh and Al-Berzan, 2007; Krstić et al., 2007; Yahong et al., 2006; Xinwei et al., 2006; Righi and Bruzzi, 2006; Pavlidou et al., 2006; Walley El-Dine et al., 2001). Although a few studies related to the radioactivity in granite tiles were published in literature (Örgün et al., 2005; Turhan et al., 2008; Canbaz et al., 2010; Karadeniz et al., 2011) the detailed information of possible radiological hazards from the natural radioactivity in commercially-utilized ornamental and countertops granite tiles in Turkey is not available in the literature.

In the present study, the estimation of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in granite tile samples was carried out by using gamma-ray spectrometer with high purity germanium (HPGe) detector. Potential radiological hazards caused from the utilization of these materials in homes were evaluated by calculating external exposure index (activity concentration index), internal exposure index (alpha index) and indoor absorbed gamma dose rate arose from external exposure, taking into account European Commission (EC) guideline (EC, 1999), and national standard (TS 12614, 1999) and international report (UNSCEAR, 2000).

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2. Materials and method

2.1. Sample preparation for radiometric measurements

A total of 42 different granite tile samples were collected from local major retailers, cataloged and coded properly. All samples were pulverized and then dried in a temperature-controlled furnace at 115 °C for 10–15 h to remove moisture. Each sample was then transferred to plastic containers. The weighed containers were hermetically sealed and stored for more than 30 days to capture ^{222}Rn gas and ensure secular equilibrium between ^{226}Ra and its decay products as well ^{228}Ra and its short lived decay products. Furthermore, an assumption of radioactive equilibrium between ^{238}U and ^{226}Ra, and between ^{232}Th and ^{228}Th was made. The geometrical dimensions of the samples were kept identical to those of the reference materials.

2.2. Instrumentation and calibration

The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in granite samples were measured using a high-resolution gamma-ray spectrometric system. The system was consisted of a coaxial p-type HPGe detector which was shield to reduce background. The detector was interfaced to the digital spectrum analyzer which was a full-featured 16 K channel multichannel analyzer on advanced digital signal processing techniques.

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 using the gamma-rays of 352 keV from ^{214}Pb and 609 keV from ^{214}B, while the gamma-rays of the 911 keV from ^{228}Ac and 583 keV from ^{208}Tl were used to determine the activity concentration of ^{232}Th. The activity concentration of ^{40}K was measured directly by its gamma-ray at 1461 keV. The sample containers were placed on top of the detector for counting (close geometry). The same geometry was used to determine peak area of samples and references. Measurement time for each was sufficiently long to provide that uncertainty in the counting rates was less than 10% at the 95% confidence level. Prior to sample measurement gamma-ray background at the laboratory was determined with an empty container under the same conditions of sample measurements and subtracted in order to get net counts for the sample.

The activity concentration (A) of radionuclides mentioned above was calculated from the following equation:

$$A(\mathrm{Bq\ kg^{-1}}) = \frac{C_\mathrm{N}}{\varepsilon(E_\gamma)\cdot I_\gamma \cdot t \cdot M} \tag{1}$$

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

The absolute efficiency calibration of the gamma spectrometry systems was carried out using the radionuclide specific efficiency method in which the efficiency values of gamma-ray lines belonging to the specific radionuclide existing only in both the reference material and sample were used. Thus, the uncertainty in gamma-ray intensities, the influence of coincidence summation and self-absorption effects of the emitting gamma photons were avoided for the close geometry conditions. The reference materials RGU-1 (U-ore), RGTh-1 (Th-ore), RGK-1 (K_2SO_4) and soil 375 obtained from International Atomic Energy Agency were employed for the radionuclide specific efficiency calibration of the counting system. The measured efficiency values obtained for close geometry were fitted to the third order polynomial as follows:

$$\varepsilon(E_\gamma) = \exp\left(\sum_{i=0}^{3} a_i (\ln E_\gamma)^i\right) \tag{2}$$

where E_γ is gamma-ray energy and a_i-s are fitted parameters (Fig. 1).

3. Results and discussion

3.1. Estimation of natural radioactivity

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the examined granite tile samples are shown in Table 1. As can be seen from Table 1, the highest values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K measured in granite tile samples are 192 ± 6 (GT4), 345 ± 10 (GT14) and 4156 ± 393 (GT32) Bq kg^{-1}, respectively, while the lowest values of the activity concentration of the same radionuclides are 9.2 ± 0.5 (GT37), 7.5 ± 0.4 (GT12) and 92 ± 6 (GT38) Bq kg^{-1}, respectively. In Table 2, the average values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K are compared with the corresponding values measured in other countries and

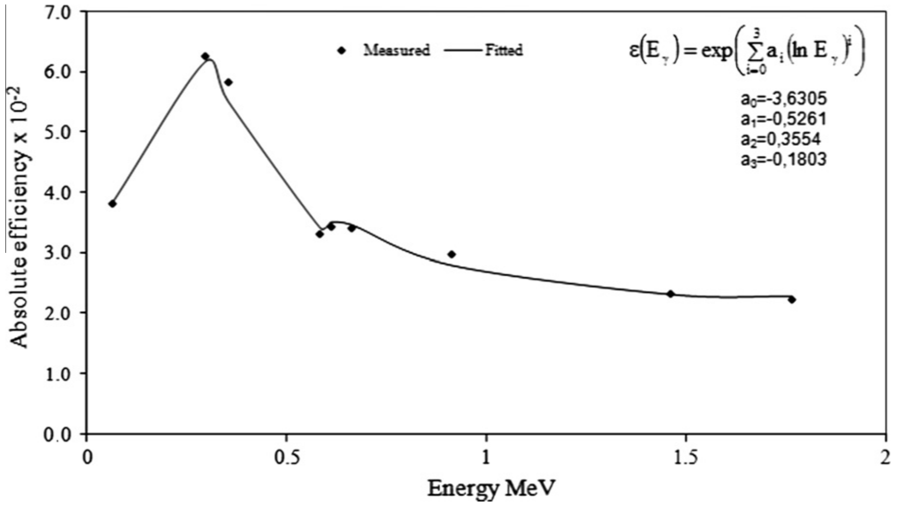

Fig. 1. Absolute efficiency curve of the HPGe detector for counting geometries used in this study.

Table 1
The values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K obtained for the 42 different granite tiles.

Sample code	Activity concentration (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
GT1	131 ± 7	101 ± 5	1310 ± 126
GT2	70 ± 32	86 ± 3	1005 ± 90
GT3	147 ± 4	109 ± 5	1175 ± 89
GT4	193 ± 6	226 ± 10	1925 ± 160
GT5	67 ± 2	64 ± 3	628 ± 59
GT6	90 ± 3	97 ± 4	780 ± 71
GT7	148 ± 6	146 ± 6	1478 ± 140
GT8	117 ± 3	150 ± 5	1313 ± 128
GT9	61 ± 2	59 ± 3	1149 ± 105
GT10	111 ± 5	130 ± 5	1264 ± 115
GT11	98 ± 4	119 ± 4	1458 ± 135
GT12	17.8 ± 0.5	7.5 ± 0.4	162 ± 16
GT13	23.8 ± 0.7	29.0 ± 1.2	496 ± 48
GT14	80 ± 2	345 ± 10	1290 ± 111
GT15	128 ± 5	109 ± 5	1250 ± 105
GT16	22.6 ± 0.5	104 ± 4	1195 ± 91
GT17	49.9 ± 2.5	53 ± 2	676 ± 47
GT18	125.9 ± 7.7	150 ± 5	1330 ± 112
GT19	129.0 ± 7.4	131 ± 4	1276 ± 106
GT20	164.8 ± 6.6	144 ± 6	1792 ± 156
GT21	77.9 ± 2.3	74 ± 3	915 ± 63
GT22	95.4 ± 2.6	76 ± 3	1005 ± 80
GT23	142.4 ± 7.8	115 ± 5	1501 ± 150
GT24	74.5 ± 3.5	57 ± 2	153 ± 16
GT25	65.3 ± 2.8	122 ± 5	1317 ± 140
GT26	127.3 ± 5.4	96 ± 4	378 ± 29
GT27	64.9 ± 3.2	81 ± 4	963 ± 79
GT28	58.8 ± 3.1	78 ± 4	845 ± 71
GT29	82.4 ± 4.4	85 ± 4	795 ± 69
GT30	14.4 ± 0.8	58 ± 2	1209 ± 135
GT31	65.4 ± 2.3	122 ± 5	1317 ± 91
GT32	134.2 ± 6.7	208 ± 10	4156 ± 393
GT33	45.0 ± 1.9	36.9 ± 1.6	283 ± 26
GT34	105 ± 5	52 ± 3	587 ± 53
GT35	112 ± 6	170 ± 7	1106 ± 101
GT36	24.7 ± 1.2	39.1 ± 1.9	1101 ± 107
GT37	9.2 ± 0.5	60 ± 3	1624 ± 154
GT38	18.7 ± 1.0	14.1 ± 0.7	92 ± 6
GT39	91 ± 5	18.2 ± 0.9	552 ± 44
GT40	103 ± 6	13.9 ± 0.7	530 ± 38
GT41	115 ± 7	15.0 ± 0.6	502 ± 39
GT42	114 ± 6	52 ± 2	436 ± 34
Average ± SE	88 ± 7	95 ± 10	1055 ± 103

the average values of natural radionuclides in earth's crust. From Table 2, it is concluded that the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in granite tile samples from different areas in the world vary over a wide range. The average concentration values of ^{226}Ra and ^{40}K in the granite tile samples is about three times higher than the earth's crust average values of 32 and 420 Bq kg^{-1} for ^{226}Ra and ^{40}K, respectively, while the average concentration value of ^{232}Th is two times higher than the earth's crust average value of 45 Bq kg^{-1}. The average concentration values of these radionuclides in granite tile samples are about one and half times higher than typical concentration values of 60, 60 and 640 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively, measured in natural building stones utilized in the European Union (EU) countries (EC, 1999). Also, the average values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K obtained in this study are similar to the world wide averages of 42, 73 and 1055 Bq kg^{-1}, respectively.

3.2. Evaluation of possible radiation hazards caused from exposure due to radionuclides

The external exposure index (activity concentration index), internal exposure index (alpha index) and indoor absorbed gamma dose rate were determined to evaluate the radiation hazards associated with the usage of the granite tile materials samples examined.

3.2.1. External exposure index (activity concentration index)

A number of indices related to the evaluation of the external exposure to gamma radiation arising from building materials have been proposed by several investigators (Righi and Bruzzi, 2006). In the present study, the activity concentration index (ACI) proposed by the European Commission (EC) recommendation was taken into consideration because more than one radionuclide contribute to the radiation dose (EC, 1999). ACI has been used for practical controlling purposes. In EU countries, building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at most (EC, 1999). As well as the criterion for controlling was established considering the overall nations circumstances, it is recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv. ACI was calculated for building using the following formula (EC, 1999):

$$\mathrm{ACI} = \frac{A_{\mathrm{Ra}}}{300\ \mathrm{Bq\ kg^{-1}}} + \frac{A_{\mathrm{Th}}}{200\ \mathrm{Bq\ kg^{-1}}} + \frac{A_{\mathrm{K}}}{3000\ \mathrm{Bq\ kg^{-1}}} \tag{3}$$

where A_{Ra}, A_{Th} and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in terms of Bq kg^{-1}, respectively. For superficial materials (tiles and natural building stones), ACI $\leqslant$ 2 corresponds to an annual

Table 2
Average values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in granite samples from the different countries and earth crust.

Country	Activity concentration (Bq kg^{-1})			References
	^{226}Ra	^{232}Th	^{40}K	
Brasil	31	73	1648	Anjos et al. (2005)
China	88	114	1270	Xinwei et al. (2006)
China (average)	90	116	969	Yahong et al. (2006)
Egypt (Wadi Karim)	56	54	4849	El-Arabi (2007)
Greece	64	81	104	Pavlidou et al. (2006)
USA	57	69	1140	Kitto et al. (2009)
Pakistan	51	70	1853	Amanat et al. (2002)
Pakistan	659	598	1203	Asghar et al. (2008)
Turkey (Sivrihisar)	67[a]	153	1058	Örgün et al. (2005)
Turkey (Ezine)	175	205	1172	Örgün et al. (2007)
Turkey	88	95	1055	This study
Yemen	54	127	1743	Abd El-mageed et al. (2011)
Earth crust average	32	45	420	UNSCEAR (2000)
Natural building stones (European Union)	60	60	640	EC (1999)
Worldwide	42[a]	73	1055	Kitto et al. (2009)

[a] This value is given for ^{238}U.

Table 3
The values of the activity concentration index (ACI), the alpha index (AI) and the indoor absorbed gamma dose rate (D_R) evaluated for the samples of the granite tiles.

Sample code	ACI	AI	D_R (nGy h^{-1})	E (mSv)
GRAT1	1.38	0.66	42	0.21
GRAT2	1.00	0.35	30	0.15
GRAT3	1.43	0.74	44	0.22
GRAT4	2.41	0.96	73	0.36
GRAT5	0.75	0.33	23	0.11
GRAT6	1.04	0.45	32	0.16
GRAT7	1.71	0.74	52	0.26
GRAT8	1.58	0.58	48	0.23
GRAT9	0.88	0.30	27	0.13
GRAT10	1.44	0.55	44	0.21
GRAT11	1.41	0.49	42	0.21
GRAT12	0.15	0.09	5	0.02
GRAT13	0.39	0.12	12	0.06
GRAT14	2.42	0.40	70	0.34
GRAT15	1.39	0.64	43	0.21
GRAT16	0.99	0.11	29	0.14
GRAT17	0.66	0.25	20	0.10
GRAT18	1.61	0.63	49	0.24
GRAT19	1.51	0.64	46	0.23
GRAT20	1.87	0.82	57	0.28
GRAT21	0.93	0.39	28	0.14
GRAT22	1.03	0.48	32	0.16
GRAT23	1.55	0.71	48	0.23
GRAT24	0.58	0.37	18	0.09
GRAT25	1.27	0.33	38	0.18
GRAT26	1.03	0.64	32	0.16
GRAT27	0.94	0.32	28	0.14
GRAT28	0.87	0.29	26	0.13
GRAT29	0.97	0.41	29	0.14
GRAT30	0.74	0.07	21	0.11
GRAT31	1.27	0.33	38	0.18
GRAT32	2.87	0.67	85	0.42
GRAT33	0.43	0.22	13	0.07
GRAT34	0.81	0.52	26	0.13
GRAT35	1.59	0.56	48	0.23
GRAT36	0.64	0.12	19	0.09
GRAT37	0.87	0.05	25	0.12
GRAT38	0.16	0.09	5	0.03
GRAT39	0.58	0.45	19	0.09
GRAT40	0.59	0.51	19	0.09
GRAT41	0.63	0.57	21	0.10
GRAT42	0.79	0.57	25	0.12
Average ± SE	1.12 ± 0.09	0.44 ± 0.03	34 ± 3	0.17 ± 0.01

effective dose less than or equal 0.3 mSv, while ACI $\leqslant 6$ corresponds to an annual effective dose less than or equal 1 mSv.

The values of the calculated ACI for granite tile samples are shown in the second column of Table 3 and Fig 2a. From Table 3, the values of the ACI varied from 0.15 to 2.87 with a mean of 1.12 ± 0.09. It is observed in Fig. 2a that all values of ACI for the examined granite tile samples are significantly below the criterion of 6 corresponding to an effective dose 1 mSv. Furthermore, the values of ACI are lower than the criterion of 2 corresponding to an effective dose 0.3 mSv, except for GT4, GT14 and GT32.

3.2.2. 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 (Righi and Bruzzi, 2006). In this study, the alpha index (AI) was calculated by the next formula:

$$\mathrm{AI} = \frac{A_{\mathrm{Ra}}}{200\ \mathrm{Bq\ kg^{-1}}} \tag{4}$$

where A_{Ra} is the activity concentration of ^{226}Ra 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} (TS 12614, 1999). AI $\leqslant 1$ corresponds to the ^{226}Ra activity concentration less than or equal 200 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 Bq m^{-3}.

The values of the calculated AI for granite tile samples are shown in the third column of Table 3 and Fig 2b. From Table 3, the values of the AI varied from 0.05 to 0.96 with a mean of 0.44 ± 0.03. It is seen from Fig. 2b that all values of AI for the examined granite tile samples are below the recommended upper level of unity.

3.2.3. Indoor absorbed gamma dose rate

It is possible to evaluate the indoor absorbed dose rate due to the external gamma radiation arising from the granite tile samples utilized in construction of dwellings and work places from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured for each sample. The indoor absorbed gamma dose rate (D_{in}) in air was evaluated using data and formulae provided by the EC report (EC, 1999). In the EC report, the dose conversion coefficients were calculated for the center of the room. The dimensions of the room are 4 m × 5 m × 2.8 m. 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 nGy h^{-1} per Bq kg^{-1} for ^{40}K:

$$D_{\mathrm{in}}(\mathrm{nGy\ h^{-1}}) = 0.12 \times A_{\mathrm{Ra}} + 0.14 \times A_{\mathrm{Th}} + 0.0096 \times A_{\mathrm{K}} \tag{5}$$

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}.

The results of the D_{in} evaluated for the samples of the granite tile samples are given in the fourth column of Table 3. The evaluated values of D_{in} for the samples examined varied from 5 to 85 nGy h^{-1} with a mean of 34 nGy h^{-1}. From the data in Table 3, all evaluated values of D_{in} for the samples examined, except for one granite sample (85 nGy h^{-1} for GT32) are lower than the world average (populated-weighted) indoor absorbed gamma dose rate of 84 nGy h^{-1} (UNSCEAR, 2000). The relative contributions to total absorbed indoor gamma dose rates due to ^{226}Ra and ^{232}Th decay products and ^{40}K for commercial granite samples are 33%, 37% and 30%, respectively.

3.2.4. Indoor annual effective dose

The indoor annual effective dose (E_{in}) was estimated using the following formula:

$$E_{\mathrm{in}}(\mathrm{mSv}) = D_{\mathrm{in}}(\mathrm{nGy\ h^{-1}}) \times F_{\mathrm{C}}(\mathrm{Sv\ Gy^{-1}}) \times T_{\mathrm{E}}(\mathrm{h\ y^{-1}}) \times 10^{-6} \tag{6}$$

where D_{in} is the indoor absorbed gamma dose rate, F_{C} is the conversion factor of 0.7 Sv Gy^{-1} from indoor absorbed gamma dose in air to effective dose received by adults (UNSCEAR, 2000) and T_{E} is annual exposure time of 7000 h y^{-1}, implying that 80% of time is spent indoors.

The estimated results for the annual effective dose (E_{in}) are given in the fifth column of Table 3 for the granite tile samples. The values of E_{in} varied from 0.02 to 0.42 with a mean of 0.17 ± 0.01. It can be seen from Table 3 that all values of E_{in}, except for one granite sample (0.42 mSv for GT32) are significantly lower than the exemption dose criterion of 0.3 mSv.

4. Conclusions

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K terrestrial radionuclides in the granite tile samples commercially utilized as ornamental and countertops in a home in Turkey were estimated using high resolution gamma-ray spectrometry with the HPGe detector. The results show that the average concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the granite tile samples are signif-

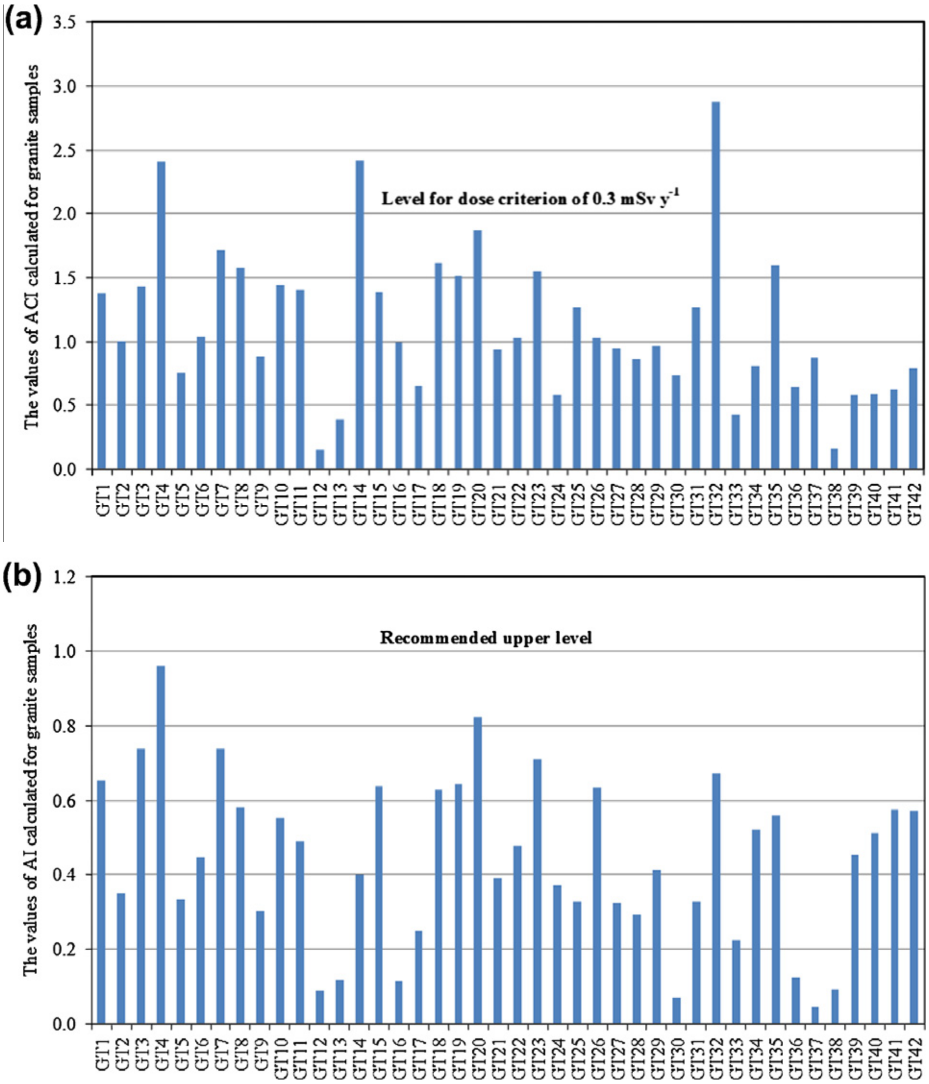

Fig. 2. Calculated values of external (a) and internal (b) exposure index for the examined commercial granite samples.

icantly higher than the earth's crust averages. Turkey has adopted the radiological protection principles concerning the natural radioactivity of building materials developed by the European Commission. It is recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv (control level). For each granite sample the activity concentration index, the alpha index, the indoor absorbed gamma dose rate and the corresponding annual effective dose were determined to evaluate the possible radiological hazard for exposure of the occupants from the utilization of the samples as ornamental and countertops in construction sector. The results of activity concentration index show that all granite samples with the exception of three samples meet the exemption annual dose criterion of 0.3 mSv. The values of alpha index calculated for the samples were found well below the acceptable limit of 1. The indoor gamma dose rates calculated for the samples are lower than the population-weighted average of 84 nGy h^{-1} exception for one granite tile Also, the values of the corresponding annual effective dose are significantly lower than the dose criterion of 1 mSv.

As a conclusion, the present study indicates that the dose due to the utilization of the granite tile samples examined are within the recommended values and do not pose any significant source of radiation hazard.

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