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ARTICLE

Radiometric analysis of volcanic tuff stones used as ornamental and structural building materials in Turkey and evaluation of radiological risk

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Abstract – Volcanic tuff stones are widely used to coat the interior and exterior surfaces of buildings for ornamental purposes in the construction industry in Turkey, and are also used as structural material in the construction of masonry buildings, especially in the Cappadocia region, which is a popular tourist destination. In this study, seventy-six volcanic tuff stone samples collected from different quarries located in different geographical regions in Turkey were surveyed for radiometric analysis, and the radiological risk to human health caused by ionizing radiation from natural radionuclides in the volcanic tuff stone samples was evaluated. The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K analyzed in volcanic tuff stone samples were very variable, ranging from 3 to 389, 8 to 401, 2 to 263 and 99 to 2107 Bq kg^{-1}, respectively. The emanation coefficient and mass exhalation rate of ^{222}Rn measured for the volcanic tuff samples ranged from 2 to 68% with an average of 34%, and 1 to 194 μBq kg^{-1} s^{-1} with an average of 43 μBq kg^{-1} s^{-1}, respectively. The external (gamma) and internal (alpha) hazard indices, indoor absorbed gamma dose rate and the corresponding annual effective dose were estimated to evaluate the potential radiological risk to human health. The results showed that all of the surveyed volcanic tuff stone samples can be used as covering building materials for ornamental or insulating covering purposes. However, the use of some volcanic tuff stone samples as structural building materials should be restricted.

Keywords: natural radioactivity / radon mass exhalation rate / emanation coefficient / hazard indices / annual effective dose / radiological risk / volcanic tuff

1 Introduction

All building raw materials and products derived from rock and soil contain various amounts of naturally occurring radionuclides of the uranium-radium (^{238}U-^{226}Ra) and thorium (^{232}Th) series, and the radioactive isotopes of potassium (^{40}K). Enhanced or elevated levels of natural radionuclides in building materials may cause a radiological risk to human health due to external and internal exposure (Marocchi *et al.*, 2011). The external exposure is caused by direct gamma radiation originating from the above-mentioned members of radioactive series, while internal exposure is caused by the inhalation of the radioactive inert gas radon (^{222}Rn, a daughter product of ^{226}Ra) and its short-lived secondary decay products, which are exhaled from building materials into room air. Knowledge of the level of natural radioactivity in building materials is therefore essential to evaluate the possible radiological hazards to human health and develop standards and guidelines for the use and management of these materials (Turhan, 2009).

Volcanic tuff stones consisting of consolidated volcanic ash, and large and small pieces ejected from vents during a volcanic eruption, with a porous structure in different colours, are used as covering materials for insulating and ornamental purposes on the exterior and interior of buildings in Turkey. Up until now, several studies on the radiometric, petrographic and mineralogical properties of natural stone samples have been reported (Anjos *et al.*, 2005; Xinwei *et al.*, 2006; El-Arabi, 2007; Asghar *et al.*, 2008; Kitto *et al.*, 2009; Koornneef *et al.*, 2010; Lanzo *et al.*, 2010; Moura *et al.*, 2011; Abd El-Ghafour *et al.*, 2012; Capaccioni *et al.*, 2012; Trevisi *et al.*, 2012; Turhan, 2012). However, according to our literature survey, a comprehensive study related to the radiometric properties of volcanic tuff samples collected from Turkey has not yet been conducted, except one study in which eight volcanic tuff samples were considered (Değerlier, 2013). In this study, a total of 76 volcanic tuff stones collected from different quarries located in four geographical regions of Turkey were analyzed using gamma-ray spectrometry to determine the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K, and the emanation

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Table 1. Information on the locations of quarries, sample codes and sample colors.

Quarry code	N°	Sample colors (number of samples)	Quarry place (geographical region)
Q1	6	Yellow (2), black (1), cherry (1), dark gray (1), dusty-rose (1)	Kayseri-Tomarza (Central Anatolia)
Q2	6	Yellow (2), brown (1), black (1), dusty-rose (2)	Kayseri-Tomarza (Central Anatolia)
Q3	7	Cherry (1), red (1), brown (1), yellow (1), gray (1), dusty-rose (1), black (1)	Kayseri-Tomarza (Central Anatolia)
Q4	7	Gray (4), black (1), dusty-rose (1), brown (1)	Kayseri-Tomarza (Central Anatolia)
Q5	4	Yellow (3), brown (1)	Kayseri-Tomarza (Central Anatolia)
Q6	6	Yellow (1), black (1), gray (1), dusty-rose (1), fawn (1), dark brown (1)	Kayseri-Develi (Central Anatolia)
Q7	4	Gray (4)	Kayseri-Melikgazi (Central Anatolia)
Q8	3	Gray (2), yellow (1)	Kayseri (Central Anatolia)
Q9	6	Yellow (1), cherry (1), black (1), white (1), red (1), turquoise (1)	Nevşehir-Ürgüp (Central Anatolia)
Q10	5	Yellow (1), cherry (1), dusty-rose (1), light brown (1), black (1)	Nevşehir-Avanos road (Central Anatolia)
Q11	4	Yellow-white (1), white (1), dark yellow (1), Cappadocia rose (1)	Nevşehir-Aksaray road (Central Anatolia)
Q12	1	Brown (1)	Nevşehir-Ürgüp (Central Anatolia)
Q13	6	Fawn (2), dark brown (1), beige (1), brown (1), cream (1)	Isparta (Mediterranean)
Q14	6	Beige (1), yellow (1), gray (1), cream (1), dusty-rose (1), white (1)	Afyon (Aegean)
Q15	2	Brown (2)	Manisa (Aegean)
Q16	2	Rose-colored (2)	Eskişehir (Central Anatolia)
Q17	1	Maroon (1)	Diyarbakır (Southeast Anatolia)

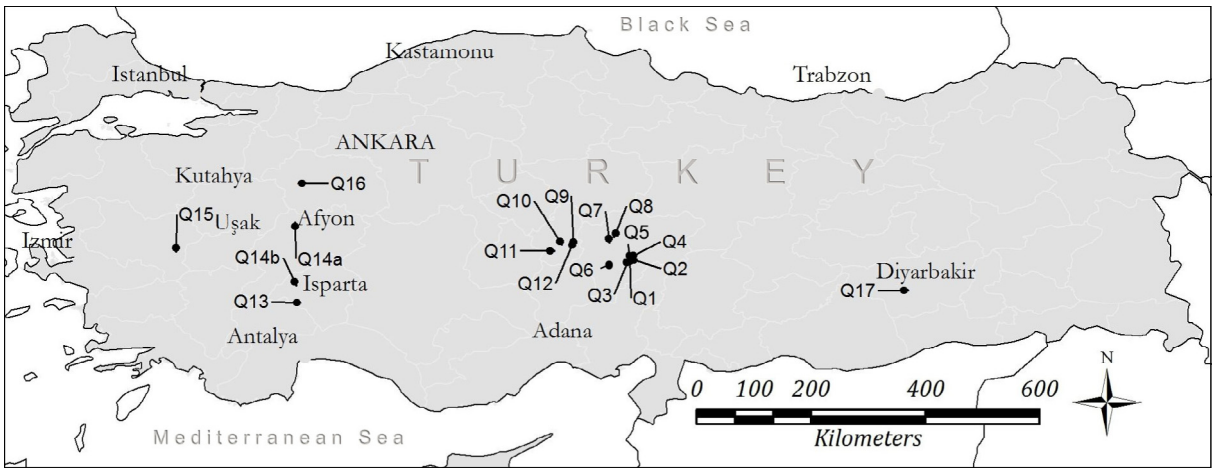

Figure 1. Map showing the volcanic tuff samples' locations.

coefficient and mass exhalation rate of ^{222}Rn. The gamma index, alpha index, indoor absorbed gamma dose rate and the corresponding annual effective dose were estimated for each volcanic tuff stone to evaluate the potential radiological risk to members of the public, spending most of their time indoors. The results obtained in this study were compared with the upper or reference values recommended or proposed by the World Health Organization (WHO), United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and European Commission (EC).

2 Materials and method

2.1 Sampling and sample preparation

Volcanic tuff stone samples were collected from different quarries located in Central Anatolia, Eastern Anatolia, and Mediterranean and Aegean regions in Turkey, catalogued and coded properly (Table 1). The sampling locations in the study areas are shown in Figure 1. The samples were crushed, pulverized and then dried in a temperature-controlled furnace at 105 °C for 24 h to remove moisture. After moisture removal, these samples were cooled in a moisture-free atmosphere. Each sample was transferred to a 1-L Marinelli container and weighed. Sample containers were hermetically sealed and allowed to stand for at least four weeks to ensure short-term equilibrium between ^{226}Ra and its short-lived decay products.

2.2 Measurement system for radiometric analysis

Radiometric analyses were performed using a gamma-ray detection system with a high-resolution coaxial p-type horizontal HPGe detector (Canberra GX3018). The resolution of the detector is 1.8 keV for ^{60}Co gamma-ray line energy at 1332.5 keV and it has a relative efficiency of 30%. The detector

was shielded to minimize natural background radiation from the environment. The certificated standard calibration source of a 1-L Marinelli beaker which contains multinuclides distributed in 1.0 g cm^{-3} epoxy (Eckert & Ziegler Isotope Products) was used for efficient calibration of the system in the energy range from 122 keV to 1836 keV. The counting time for each sample was adjusted to obtain a gamma-ray spectrum with good statistics. The activity concentration of ^{238}U was derived from the weighted average of the gamma-ray lines of 63.3 keV and 92.3 keV of ^{234}Th corrected for self-adsorption effects. The activity concentration of ^{40}K was measured directly by its own gamma ray at 1460.8 keV, while the activities of ^{226}Ra and ^{232}Th were calculated based on the weighted mean value of their respective decay products in secular equilibrium. The activity concentration of ^{226}Ra was measured using the 351.9 keV gamma-ray line from ^{214}Pb and the 609.3 keV gamma-ray line from ^{214}Bi. The activity concentration of ^{232}Th was measured using the 911.2 keV gamma-ray line from ^{228}Ac and the 583.2 keV gamma-ray line from ^{208}Tl. The lower limit of detection (L_D) of the gamma-ray spectrometer with a 95% confidence limit was estimated using the following formula (Currie, 1968):

$$L_D\left(\text{Bq kg}^{-1}\right) = \frac{1.64 \times \sigma_B}{\varepsilon_\gamma \times I_\gamma \times t \times M} \quad (1)$$

where σ_B is the standard deviation of the background in the region of interest of the gamma photopeak, ε_γ is the absolute efficiency of the system, I_γ is the gamma-ray probability per decay, t is the counting time and M is the mass of the sample. The average values of the L_D for ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in volcanic tuff stone samples were found to be 0.8, 1.2, 0.9 and 7.7 Bq kg^{-1}, respectively.

The correction for the contribution of ^{232}Th *via* its daughter nuclide ^{228}Ac (1459.2 keV peak) to the 1460.8 keV peaks of ^{40}K was made as follows (Lavi *et al.*, 2004):

$$\text{Errorin K activity}(\%) = 9.3 \times \frac{A_{Th}}{A_K} \quad (2)$$

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

3 Results and discussion

3.1 Radiometric analysis

Table 2 reports the range and average values of the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K measured in the volcanic tuff samples together with the statistical uncertainty, which was estimated by taking into consideration the systematic uncertainties in the efficiency calibration and counting statistical uncertainty. Skewness and kurtosis values and some statistical data of the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K for all the samples are given in Table 3. As can be seen in Table 2, the highest values of the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K are 389 (Q13), 401 (Q13), 263 (Q13) and 2107 (Q5) Bq kg^{-1}, respectively, while the lowest values of the activity concentration of the same radionuclides are 3 (Q1), 8 (Q1), 2 (Q1) and 99 (Q1) Bq kg^{-1}, respectively. The average activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K were found to be 94, 104, 62 and 623 Bq kg^{-1}, respectively. As can be seen in Table 3, the values of skewness and kurtosis are significantly higher than the null value. This situation signifies the non-existence of a normal situation for the radionuclides. This is also verified by the log-normal distribution shown in Figures 2a–2d. It is observed in Figure 2 that the ranges of 40–100 Bq kg^{-1}, 40–120 Bq kg^{-1}, 20–90 Bq kg^{-1} and 240–560 Bq kg^{-1} measured in tuff stone samples for ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K include 71%, 68%, 74% and 54% of the samples, respectively. The average values of these radionuclides are significantly higher than the corresponding earth crust average values, which are 33, 45, 32 and 412 Bq kg^{-1} for ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K, respectively (UNSCEAR, 2008). The typical concentration values of ^{226}Ra, ^{232}Th and ^{40}K in natural building stones utilized in European Union (EU) countries were given as 60, 60 and 640 Bq kg^{-1}, respectively (EC, 1999). The average activity concentrations of ^{226}Ra and ^{40}K are close to the corresponding EU quoted values, while the average activity concentration of ^{232}Th is about two times higher than the EU quoted value. Table 4 presents a comparison of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in this study for the volcanic tuff samples examined with those obtained for natural stone samples in other countries. As can be seen in Table 4, the values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K obtained for the volcanic tuff samples are comparable with those obtained for different natural stone samples from other studies. As can be seen in Table 2, the average activity concentrations of ^{238}U measured in the samples from 17 quarries are higher than the earth crust's average value, while the average activity concentrations of ^{232}Th, ^{226}R and ^{40}K measured in the samples from Q7, Q15 and Q17, Q7, Q10, Q11 and Q15, and Q1, Q7, Q11, Q15 and Q17, respectively, are lower than the earth crust's average value.

The activity concentration of ^{238}U, ^{232}Th and ^{40}K can be converted into the elemental concentrations of U (in mg kg^{-1}), Th (in mg kg^{-1}) and K (in percent) using the following formula:

$$C_E = \frac{A_E \times M_E \times F}{\lambda_E \times N_{Av} \times h_E} \quad (3)$$

where C_E is the elemental concentration of element E. A_E, M_E, λ_E, N_{Av} and h_E are the measured activity concentration (Bq kg^{-1}), the atomic mass (kg mol^{-1}), the decay constant (s^{-1}), Avogadro's number (6.023×10^{23} atoms mol^{-1}) and the atomic abundance in nature, respectively. F is a factor with a value of 1,000,000 for ^{238}U and ^{232}Th in mg kg^{-1} and 100 for ^{40}K in percentage. The elemental concentration of ^{238}U, ^{232}Th and ^{40}K calculated for the tuff samples ranged from 0.2 to 31.5 mg kg^{-1} with an average of 17.6 ± 0.9 mg kg^{-1}, 2.1 to 98.8 mg kg^{-1} with an average of 25.6 ± 2.5 mg $kg^{-1,}$ and from 0.3 to 7.0% with an average of 2.1 ± 0.2%, respectively. The concentrations of ^{232}Th and ^{238}U in the tuff samples were positively correlated (R = 0.81), while ^{40}K was weakly correlated with both ^{238}U and ^{232}Th concentrations (R = 0.52 and R = 0.52, respectively), as shown in Figure 3.

Table 2. The range and average value of the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K measured in the volcanic tuff samples.

Quarry code	N		Activity concentration (in Bq kg^{-1} ± 1σ)			
			^{238}U	^{232}Th	^{226}Ra	^{40}K
Q1	6	Range	3 ± 1–77± 3	8 ± 1–103 ± 3	2± 1–68 ± 2	99 ± 8–689 ± 54
		Average ± SE	45 ± 11	61 ± 14	38 ± 10	395 ± 80
Q2	6	Range	36 ± 1–117 ± 4	51 ± 2–105 ± 4	23 ± 1–81 ± 3	255 ± 20–894 ± 70
		Average ± SE	63 ± 13	67 ± 10	40 ± 9	445 ± 95
Q3	7	Range	40 ± 1–74± 3	44 ± 2–105 ± 4	25 ± 1–61 ± 2	316 ± 25–954 ± 74
		Average ± SE	58 ± 5	69 ± 8	40 ± 5	574 ± 86
Q4	7	Range	26 ± 1–89 ± 3	34 ± 1–112 ± 4.0	23 ± 1–89 ± 3	201 ± 16–872 ± 68
		Average ± SE	48 ± 10	62 ± 12	42 ± 10	415 ± 100
Q5	4	Range	130 ± 5–182 ± 5	93± 3–231± 8	67±–117± 4	385± 30–2107 ± 164
		Average ± SE	149 ± 11	150± 32	92± 11	1457 ± 382
Q6	6	Range	38 ± 1–61 ± 2	51 ± 2–233 ± 8	36 ± 1–53 ± 2	388 ± 30–693 ± 54
		Average ± SE	49 ± 4	99 ± 28	45 ± 3	506 ± 44
Q7	4	Range	26 ± 1–73 ± 3	14 ± 1–84 ± 3	8 ± 1–40± 1	137 ± 11–489 ± 38
		Average ± SE	40 ± 11	34 ± 17	19 ± 7	268 ± 83
Q8	3	Range	64 ± 2–71 ± 3	87 ± 3–91 ± 3	38 ± 1–55 ± 2	454 ± 35–534 ± 42
		Average ± SE	68 ± 2	90 ± 1	47 ± 5	491 ± 23
Q9	6	Range	46 ± 2–77 ± 3	47 ± 2–92 ± 3	23 ± 1–61 ± 2	385 ± 30–604 ± 47
		Average ± SE	56 ± 5	70 ± 7	41 ± 6	461 ± 37
Q10	5	Range	24 ± 1–67 ± 2	71 ± 3–76 ± 3	15 ± 1–41 ± 1	391 ± 31–733 ± 57
		Average ± SE	41 ± 8	74 ± 1	25 ± 5	515 ± 60
Q11	4	Range	36 ± 1–49 ± 2	69 ± 2–80 ± 3	20 ± 1–41 ± 1	120 ± 10–428 ± 33
		Average ± SE	44 ± 3	75 ± 2	30 ± 5	263 ± 67
Q12	1		103 ± 4	108 ± 4	89 ± 3	1127 ± 88
Q13	6	Range	285 ± 10–389 ± 14	306 ± 11–401 ± 14	137 ± 4–263 ± 8	1086 ± 85–1458 ± 114
		Average ± SE	346 ± 16	366 ± 15	197 ± 18	1290 ± 58
Q14	6	Range	126 ± 4–292 ± 10	1238 ± 4–194 ± 7	111 ± 4–23 ± 7	486 ± 38–1245 ± 97
		Average ± SE	211 ± 27	159 ± 10	146 ± 24	940 ± 122
Q15	2	Range	57 ± 2–60 ± 2	27 ± 1–28 ± 1.1	10 ± 1–12 ± 1	257 ± 20–305 ± 24
		Average ± SE	58 ± 1	28 ± 1	11 ± 1	281 ± 24
Q16	2	Range	75 ± 3–129 ± 5	87 ± 3–89 ± 3	27 ± 1–108 ± 3	1147 ± 89–1234 ± 97
		Average ± SE	102 ± 27	88 ± 1	67 ± 41	1193 ± 46
Q17	1		48 ± 2	17 ± 1	40 ± 1	102 ± 8

Table 3. Statistical data of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K measured in all volcanic tuff samples.

	^{238}U	^{232}Th	^{226}Ra	^{40}K
Average	94	104	62	623
Standard deviation (SD)	92	90	57	424
Standard error (SE)	11	10	7	49
Median	59	79	40	471
Min	3	8	2	99
Max	389	401	263	2107
Skewness	2.0	2.1	1.9	1.4
Kurtosis	3.0	4.1	3.1	1.6
Number of samples	76	76	76	76
Frequency distribution	Log-normal	Log-normal	Log-normal	Log-normal

3.1.1 Emanation coefficient and mass exhalation rate

The fraction of radon that can diffuse through building materials is known as the emanation coefficient (EC_{Rn}). The EC_{Rn} was determined using the following formula (White and Rood, 2001; Turhan and Gündüz, 2008):

$$EC_{Rn} = \frac{CR}{CR_o + CR} \quad (4)$$

where CR_0 and CR are the net count rate of radon at the sealing time of the sample and after equilibrium (after 4 weeks), respectively. The radon mass exhalation rate (EX_{Rn}) is the product of the emanation coefficient and ^{222}Rn production rate (Chowdhury *et al.*, 1998). The EX_{Rn} (in Bq kg^{-1} s^{-1}) was determined by the following equation:

$$EX_{Rn} = EC_{Rn} \times A_{Ra} \times \lambda_{Rn} \quad (5)$$

where EC_{Rn} is the emanation coefficient given in equation (4), A_{Ra} is the activity concentration of ^{226}Ra (in Bq kg^{-1}) and λ_{Rn} is the decay constant of ^{222}Rn (2.1×10^{-6} s^{-1}). The average

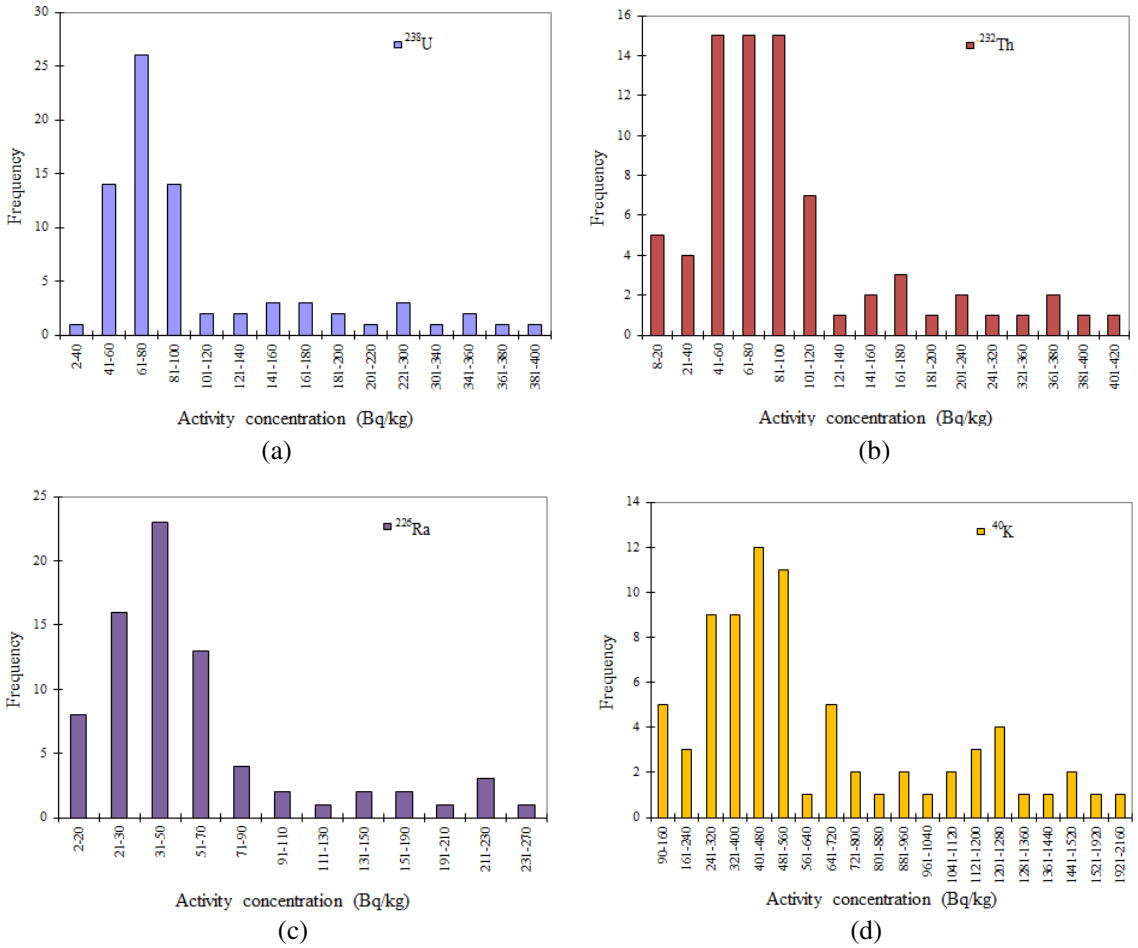

Figure 2. Frequency (number of observations) distribution of the activity concentration of ^{238}U (a), ^{232}Th (b), ^{226}Ra (c) and ^{40}K (d) in the volcanic tuff samples.

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

	N°	Country	Activity concentration (Bq kg^{-1})			References
			^{226}Ra	^{232}Th	^{40}K	
Volcanic tuff	3	Italy	92-280	137-270	1200-1900	Righi and Bruzzi, 2006
Volcanic tuff	8	Turkey (Kayseri)	18-97	17-96	229-1036	Değerlier, 2013
Volcanic	22	Italy (Vulsini)	80-394	126-487	227-2487	Capaccioni *et al.*, 2012
Volcanic tuff	76	Turkey	2-263	8-401	99-2107	This study
Ornamental stone	20	Spain	12-390	20-490	240-2000	Marocchi *et al.*, 2011
Igneous plutonic	387	EU	0.8-588	0.3-906	24-2040	Trevisi *et al.*, 2012
Igneous volcanic	86	EU	16-709	8-750	170-2354	Trevisi *et al.*, 2012
Granite	42	Turkey	9-193	8-345	92-4156	Turhan, 2012

values of the emanation coefficient and the ^{222}Rn mass exhalation rate of the volcanic tuff samples investigated in this study ranged from 2% to 68% with an average of 34%, and 1 to 194 μBq kg^{-1} s^{-1} with an average of 43 ± 5 μBq kg^{-1} s^{-1}, respectively.

3.2 Evaluation of radiological risk

In this study, the external (gamma) and internal (alpha) hazard indices, indoor absorbed gamma dose rate and the corresponding annual effective dose were estimated for each volcanic tuff sample to evaluate the radiological risk to health associated with usage of the samples as structural and ornamental building materials in Turkey.

3.2.1 Gamma index

A number of indices dealing with the evaluation of the excess gamma radiation originating from building materials

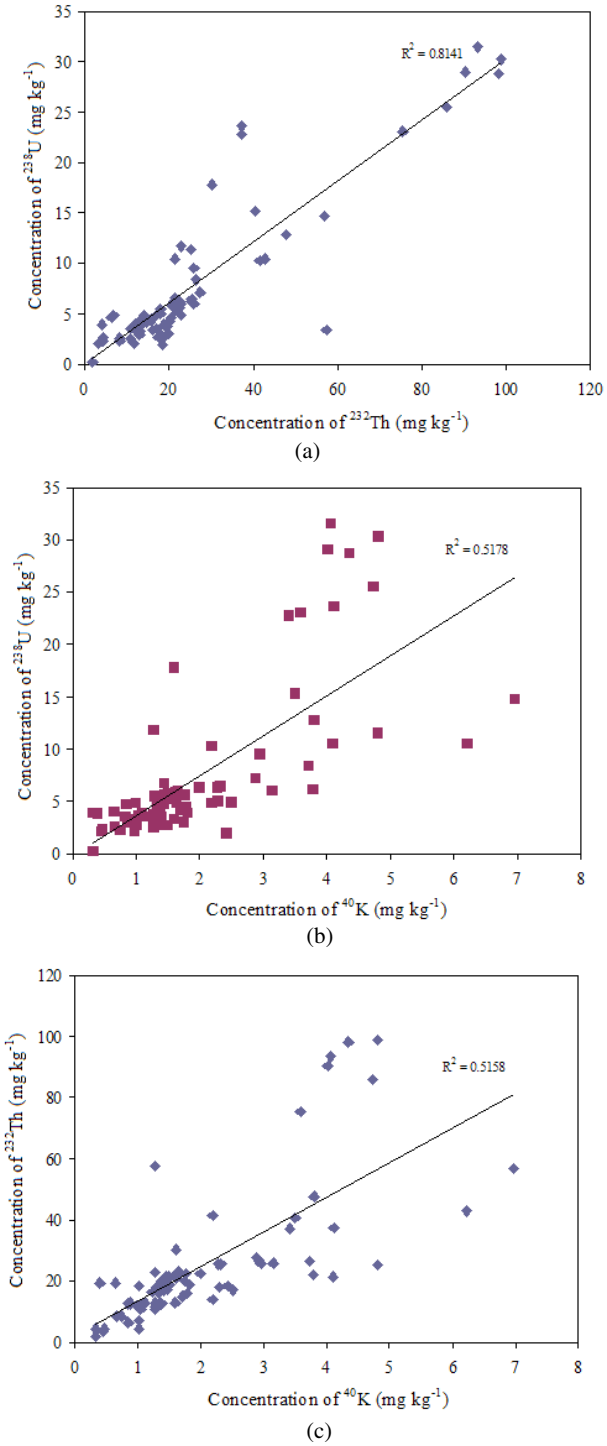

Figure 3. Correlations between ^{232}Th-^{238}U (a), ^{40}K-^{238}U (b) and ^{40}K-^{232}Th (c) in the volcanic tuff samples.

were proposed by several authors (EC, 1999). In this study, the gamma index (I_γ) proposed by the European Commission was estimated to evaluate external gamma radiation exposure caused by radionuclides in the volcanic tuff samples. In EU member 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). The criterion for controlling building materials should be established considering the overall circumstances of each nation; it is also recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv. I_γ was estimated for building materials using the following formula (EC, 1999):

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

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 structural materials used in bulk amounts, $I_\gamma \leqslant 1$ corresponds to an annual effective dose less than or equal to 1 mSv, while $I_\gamma \leqslant 0.5$ corresponds to an annual effective less than or equal to 0.3 mSv. For superficial materials, $I_\gamma \leqslant 6$ corresponds to an annual effective dose less than or equal to 1 mSv, while $I_\gamma \leqslant 2$ corresponds to an annual effective dose less than or equal to 0.3 mSv (EC, 1999). The estimated I_γ for all volcanic tuff samples ranged from 0.1 to 3.4 with an average of 0.9 ± 0.1. The average values of I_γ are given in the second column of Table 5. It can be seen in Table 5 that the highest values of the I_γ estimated for the volcanic tuff samples are 2.9 (Q13), 1.6 (Q14) and 1.5 (Q5). The average values of I_γ are below the criterion of 2 corresponding to the annual effective dose 0.3 mSv for ornamental applications, except for Q13. However, the average values of I_γ estimated for the samples from Q5, Q12, Q13, Q14 and Q16 are above the criterion of unity corresponding to the annual effective dose 1 mSv for structural applications.

Table 5. The average values of the gamma index (I_γ), alpha index (I_α), indoor absorbed gamma dose rate (D_{in}) and the corresponding annual effective dose (E_{in}).

Quarry code	I_γ	I_α	D_{in} (nGy h^{-1})	E_{in} (mSv)
Q1	0.56	0.19	133	0.6
Q2	0.61	0.20	146	0.7
Q3	0.67	0.20	158	0.8
Q4	0.59	0.21	140	0.7
Q5	1.54	0.46	366	1.8
Q6	0.81	0.23	191	0.9
Q7	0.32	0.10	76	0.4
Q8	0.77	0.23	181	0.9
Q9	0.64	0.21	152	0.7
Q10	0.62	0.12	145	0.7
Q11	0.57	0.15	132	0.6
Q12	1.21	0.45	291	1.4
Q13	2.92	0.98	687	3.3
Q14	1.59	0.73	384	1.9
Q15	0.27	0.06	63	0.3
Q16	1.06	0.34	254	1.2
Q17	0.25	0.20	64	0.3

3.2.2 Alpha index

A few internal indices dealing with the evaluation of the excess alpha radiation due to inhalation originating from

building materials were developed (Righi and Bruzzi, 2006). In this study, the alpha index (I_α) as an internal index was estimated by the following formula:

$$I_\alpha = \frac{A_{Ra}}{200 \text{ Bq kg}^{-1}} \quad (7)$$

where A_{Ra} is the activity concentration of ^{226}Ra in Bq kg^{-1}. $I_\alpha \leqslant 1$ corresponds to a ^{226}Ra activity concentration less than or equal to 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 an indoor radon concentration exceeding the recommended upper level of 200 Bq m^{-3} (Righi and Bruzzi, 2006). Recently, the WHO recommended a reference level of 100 Bq m^{-3} to minimize health hazards caused by indoor radon exposure (WHO, 2009). In this case, $I_\alpha \leqslant 0.5$ corresponds to a ^{226}Ra activity concentration less than or equal to 100 Bq kg^{-1}. The estimated I_α for all volcanic tuff samples ranged from 0.01 to 1.31 with an average of 0.31 ± 0.03. The average values of I_α are given in the third column of Table 5. It can be seen in Table 5 that all values of I_α are below the recommended upper level of unity. Also, all values of I_α are below the reference level recommended by the WHO, except for Q13 and Q14.

3.2.3 Estimation of the absorbed indoor gamma dose rate and the annual effective dose

Volcanic tuff stones are used for wall masonry and/or internal partition walls in construction of dwellings in the Cappadocia region of Turkey due to their aesthetic appearance and performance characteristics such as good heat insulation. The indoor absorbed dose rate (D_{in}) and annual effective dose (E_{in}) due to gamma-ray emission from the radionuclides in the volcanic tuff samples were evaluated using the data and formula provided by EC reports (EC, 1999). In the EC reports, the dose conversion coefficients were calculated for the standard room center. The dimensions of the room are 4 m × 5 m × 2.8 m. The thickness of the walls, floor and ceiling, and density of the structures are 20 cm and 2350 kg m^{-3} (concrete), respectively. These coefficients correspond to 0.92 nGy h^{-1} per Bq kg^{-1} for ^{226}Ra, 1.1 nGy h^{-1} per Bq kg^{-1} for ^{232}Th and 0.080 nGy h^{-1} per Bq kg^{-1} for ^{40}K.

$$D_{in}(\text{nGy h}^{-1}) = 0.92 \times A_{Ra} + 1.1 \times A_{Th} + 0.080 \times A_K \quad (8)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The conversion factor from the absorbed dose in air to the effective dose and the indoor occupancy factor has to be taken into account in order to estimate the corresponding annual effective dose. In the UNSCEAR (1982) and EC (1999) reports, a value of 0.7 Sv Gy^{-1} was used for the conversion factor from the absorbed dose in air to the effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 80% of time is spent indoors, on average, around the world (UNSCEAR, 1982). The annual effective dose (E_{in}) was estimated as follows:

$$E_{in}\,(\text{mSv}) = D_{in} \times 8766 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (9)$$

where D_{in} (nGy h^{-1}) is given by equation (8).

The estimated D_{in} values for all the volcanic tuff stone samples used as structural building materials ranged from 19 to 799 nGy h^{-1} with an average of 221 ± 20 nGy h^{-1}. The average values of D_{in} are given in the fourth column of Table 5. When the average value of 50 nGy h^{-1} for the background is subtracted from the average value of D_{in}, the remaining value of D_{in} is 2 times higher than the world average (population-weighted) indoor absorbed gamma dose rate of 84 nGy h^{-1} (UNSCEAR, 2008). It can be seen in Table 5 that the average values of D_{in} are higher than the quoted world average value, except for Q7, Q15 and Q17. The average value of D_{in} is about 3 times higher than the quoted world average value.

The estimated E_{in} values ranged from 0.1 to 3.9 mSv with an average of 1.1 ± 0.1 mSv. The average values of E_{in} are given in the fifth column of Table 5. It can be seen in Table 5 that the average values of E_{in} are lower than the dose limit value of 1, except for Q5 (1.8 mSv), Q12 (1.4 mSv), Q13 (3.3 mSv), Q14 (1.9 mSv) and Q16 (1.2 mSv). The average value of E_{in} is slightly higher than the quoted dose limit.

4 Conclusions

Radiometric analysis of 76 samples of volcanic tuff stones collected from different quarries located in Central Anatolia, Eastern Anatolia, and Mediterranean and Aegean regions in Turkey were performed using a gamma-ray spectrometer with a HPGe detector. The average activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K were measured as 94, 104, 62 and 623 Bq kg^{-1}, respectively. Also, the average values of the emanation coefficient and the ^{222}Rn mass exhalation rate of the volcanic tuff samples were found to be 68% and 43 ± 5 μBq kg^{-1} s^{-1}, respectively. The volcanic tuff samples show average concentration values that are significantly higher than the earth's crust averages.

For each sample in this study, the gamma index, alpha index, absorbed gamma dose rate in indoor air and the corresponding annual effective dose were estimated to evaluate the radiological risks. The results revealed that the radiological hazard indices estimated for all surveyed volcanic tuff stone samples do not exceed the recommended upper limit values when used as covering building materials for ornamental or insulating covering purposes. However, the Q5, Q12, Q13, Q14 and Q16 volcanic tuff samples collected from quarries could cause significant exposure to gamma radiation when used as structural materials. Therefore, the use of these volcanic tuff stone samples as structural building materials should be restricted. In case of the use of these volcanic tuff stone samples, appropriate measures such as ventilation are required.

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