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NATURAL RADIOACTIVITY, RADON EXHALATION RATES AND INDOOR RADON CONCENTRATION OF SOME GRANITE SAMPLES USED A CONSTRUCTION MATERIAL IN TURKEY

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It is very important to determine the levels of the natural radioactivity in construction materials and radon exhalation rate from these materials for assessing potential exposure risks for the residents. The present study deals with 22 different granite samples employed as decoration stones in constructions in Turkey. The natural radioactivity in granite samples was measured by gamma-ray spectrometry with an HPGe detector. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found to be in the range of 10–187, 16–354 and 104–1630 Bq kg^{-1}, respectively. The radon surface exhalation rate and the radon mass exhalation rate estimated from the measured values of ^{226}Ra content and material properties varied from 1.3 to 24.8 Bq m^{-2} h^{-1} with a mean of 10.5 ± 1.5 Bq m^{-2} h^{-1} and 0.03–0.64 Bq kg^{-1} h^{-1} with a mean of 0.27 ± 0.04 Bq kg^{-1} h^{-1}, respectively. Radon concentrations in the room caused from granite samples estimated using a mass balance equation varied from 23 to 461 Bq m^{-3} with a mean of 196 ± 27 Bq m^{-3}. Also the gamma index (I_γ), external indoor annual effective dose (E_γ) and annual effective dose due to the indoor radon exposure (E_{Rn}) were estimated as the average value of 1.1 ± 0.1, 0.16 ± 0.02 mSv and 5.0 ± 0.7 mSv, respectively, for the granite samples.

INTRODUCTION

The exposure of human beings to ionising radiation from natural sources is a continuing and inescapable feature of life on earth[1]. Natural radionuclides (cosmogenic and terrestrial) are present in various degrees in all media in the environment. Many natural rocks contain terrestrial radionuclides in the ^{238}U–^{226}Ra and ^{232}Th series and from ^{40}K. The activity concentrations of these radionuclides are related to the types of rock. Higher radiation levels are associated with igneous rocks and lower levels with sedimentary rocks[1]. External exposures in indoors and outdoors are arise mainly from gamma rays emitted from these radionuclides. Inhalation of radon (^{222}Rn) that emanates from rocks and soils and tends to concentrate in enclosed spaces such as houses is a major contributor to the internal exposure. This exposure is mostly produced by the alpha particles emitting ^{222}Rn and its short-lived daughter products. Epidemiological studies have provided convincing evidence of an association between indoor radon exposure and lung cancer, even at the relatively low radon levels commonly found in residential buildings[2]. The most common indoor radon sources are the soils and rocks under the building. However, building materials, such as concrete, bricks, natural building stones (granite, marble, etc.), natural gypsum and industrial raw materials such as fly ash, phosphogypsum, blast furnace slag, etc. may also be sources of radon emission[3]. The emanation of ^{222}Rn depends on factors such as the ^{226}Ra content in soil, rock and building materials, the material properties and the total surface area of solids, etc. This phenomenon can be explained as an exhalation process and it plays an important role in indoor and outdoor radon concentrations[4].

The knowledge of the level of natural radioactivity in building materials and the radon emissions from these materials are very important for accurately assessing the external and internal radiation exposure and developing standards and guidelines for the use and management of these materials[5]. Recently, many papers related to the natural radionuclide content of building materials were published in the literature[6–15]. Granite is the best-known igneous rock and consists mainly of quartz, mica and feldspar. It is extremely durable and scratch resistant and has different colours depending on the colour of feldspar. Granite tile is one of the most popular building materials. It has been used for thousands of years in both interior and exterior applications. Polished granite slabs and tiles are used in countertops, tile floors and many other design elements. However, the widespread use of granite tiles has caused a considerable public

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concern about the enhanced natural radiation exposure levels[15, 16]. In this study, the natural radioactivity in 22 different types of commercial granite samples used as decorative construction materials in Turkey was measured by a gamma-ray spectrometer with an HPGe detector. Radon exhalation rates (mass and surface) and the radon concentration in a typical house room with walls and floor covered with the studied granite samples were estimated by application of indirect methods. Also, the gamma index (I_γ), the external annual effective dose (E_γ) and the annual effective dose due to the indoor radon exposure (E_{Rn}) were estimated to assess potential radiological hazards to human health due to the use of these granite tile samples. The data were tabulated, discussed and compared with those given in the literature.

MATERIALS AND METHODS

Measurement of the natural radioactivity in granite samples

Granite samples collected from major retailers were ground to powder and dried. Each sample was then packed in cylindrical plastic containers and hermetically sealed. The sealed containers were stored for more than 4 weeks to ensure secular equilibrium between ^{226}Ra and its decay products as well ^{228}Ra and its short-lived decay products.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured with a gamma-ray spectrometry system equipped with a coaxial p-type HPGe detector (GC11021) with an relative efficiency of 110 % relative to a 7.62 cm (diameter) × 7.62 cm cylindrical NaI(Tl) detector, an energy resolution of 2.1 keV at 1332.5 keV of ^{60}Co and of 1.3 keV at 122 keV of ^{57}Co, and a peak-to-Compton ratio of 85:1. For gamma-ray shielding, a front opening split-top shield (Canberra Model 767) was used to reduce the background. It features lead of 10-cm thickness, which is jacketed by a 9.5-mm steel outer housing. The graded liner comprises 1-mm thickness of tin layer and 1.5-mm thickness of copper layer to prevent interference by lead X-rays. To minimise scattered radiation from the shield, the detector was centred in it. The detector was interfaced to the digital spectrum analyzer (DSA-1000), which was a full-featured 16K 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.[5].

Energy calibration of the detector was carried out using point sources (^{60}Co, ^{137}Cs and ^{241}Am). The absolute efficiency calibration of the gamma spectrometry systems 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 summation and self-absorption effects of the emitting gamma photons were avoided for the close geometry conditions. The IAEA reference materials RGU-1 (U-ore), RGTh-1 (Th-ore) and RGK-1 (K_2SO_4), with densities similar to the samples to be measured after pulverisation, were employed for the efficiency calibration of the system[5]. Each sample was counted for ~12 h. Background measurements were subtracted in order to obtain net counts for the sample.

The activity concentration of ^{226}Ra was determined from the gamma-ray line of 352 keV from ^{214}Pb, while the gamma-ray line of 583 keV from ^{208}Tl were used to determine the activity concentration of ^{232}Th. The activity concentration of ^{40}K was obtained using its 1461-keV gamma-ray line.

The minimum detectable activity (MDA, Bq kg^{-1}) of the gamma-ray measurements in 95 % confidence level was calculated using the following formula[5,17]:

$$\mathrm{MDA} = \frac{1.64 \cdot \sigma}{\varepsilon \cdot I_\gamma \cdot t \cdot M} \tag{1}$$

where σ 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, ε the absolute efficiency of the detector, I_γ the absolute emission probability of the gamma decay, t the measurement time in seconds and M the weight of the dried sample expressed in kg. The mean values of MDA for ^{226}Ra, ^{232}Th and ^{40}K were estimated to be 0.4, 0.6 and 2.7 Bq kg^{-1} for the granite samples, respectively.

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

$$\frac{\Delta A}{A} = \sqrt{\left(\frac{\Delta N_R}{N_R}\right)^2 + \left(\frac{\Delta I_\gamma}{I_\gamma}\right)^2 + \left(\frac{\Delta \varepsilon}{\varepsilon}\right)^2 + \left(\frac{\Delta M}{M}\right)^2} \tag{2}$$

where ΔN_R is the count rate uncertainty, ΔI_γ the emission probability uncertainty found in the nuclear data tables, $\Delta\varepsilon$ the efficiency uncertainty and ΔM the weighing uncertainty.

Radon exhalation rate

It is well known that a fraction of ^{222}Rn produced within the grains of rocks, soils and building materials by the radioactive decay of ^{226}Ra can escape to the pore space. The rate at which ^{222}Rn escapes from solid into the surrounding air is called an exhalation rate of the solid[18]. The exhalation rate can be

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defined as the number of radon isotope atoms that left the unit surface area of the solid material per unit time (called the surface exhalation rate) or as the number of radon isotope atoms that left the unit mass of the solid material per unit time (called the mass exhalation rate). Although ^{222}Rn exhalation mainly depends on the content of its parent radionuclide ^{226}Ra in the solid material, it is influenced by material properties and environmental factors such as humidity and temperature.

The knowledge of the exhalation rates of the building materials is helpful to study radon health hazard because some building materials may be responsible for increased indoor radon concentrations. In this study, the radon surface exhalation rate (EXR_S, Bq m^{-2} h^{-1}) was theoretically estimated by the following equation for dry conditions[16]:

$$EXR_S = \frac{1}{2} A_{Ra} \cdot \lambda \cdot \rho \cdot \eta \cdot d \quad (3)$$

where A_{Ra} is the activity concentration (Bq kg^{-1}) of ^{226}Ra, λ the decay constant of ^{222}Rn (7.6×10^{-3} h^{-1}), ρ the material density (kg m^{-3}), η the emanation coefficient and d the wall thickness (m). The radon mass exhalation rate (EXR_M, Bq kg^{-1} h^{-1}) was estimated using the following equation for dry conditions[19]:

$$EXR_M = A_{Ra} \cdot \lambda \cdot \eta \quad (4)$$

where A_{Ra} is the activity concentration (Bq kg^{-1}) of ^{226}Ra, λ the decay constant of ^{222}Rn (7.6×10^{-3} h^{-1}) and η the emanation coefficient.

Indoor radon concentration

The time-behaviour of the indoor radon concentration can be described by the mass–balance between the entry rate and the removal rate because of being radon a noble gas. The accumulation of radon in a room depends on four factors: room volume, ventilation rate, radon exhalation rate of the material and radon decay constant. Under steady conditions, the radon concentration (Bq m^{-3}) in a room due to exhalation from the indoor material can be given by[20]

$$C_{Rn} = \frac{EXR_S \cdot S}{(\lambda + \lambda_V) \cdot V} \quad (5)$$

where EXR_S is the radon surface exhalation rate (Bq m^{-2} h^{-1}), S the exhaling surface area (m^2), λ the decay constant of ^{222}Rn, λ_V the air removal rate due to ventilation (h^{-1}) and V the air volume of the room (m^3).

Gamma index

Gamma index (I_γ) was estimated to assess the radiological hazards of the potential use of granite samples used as decorative or covering building materials using the following formula[3]:

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

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. The I_γ should be used only as a screening tool for identifying materials which might be of concern. For superficial and other materials with restricted use, e.g. tiles $I_\gamma \leq 2$ corresponds to an annual effective dose $\leq$0.3 mSv, while $I_\gamma \leq 6$ corresponds to an annual effective dose $\leq$1 mSv[3].

External annual effective dose

In a house, the external annual effective dose caused by the indoor exposure depends on the activity concentrations of the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the building materials, the properties of these materials (density, thickness and elemental composition) and the type of the house[16]. The external annual effective dose (E_γ, mSv) was estimated by taking into account the indoor absorbed gamma dose rate, conversion factor and indoor occupancy factor as follows:

$$E_\gamma = D_{R\gamma}(\text{nGy h}^{-1}) \cdot 8766\ \text{h y}^{-1} \cdot O_F \cdot C_F \cdot 10^{-6} \quad (7)$$

where $D_{R\gamma}$ is the indoor absorbed gamma dose rate, O_F the indoor occupancy factor taken as 0.8 implying that 80 % of time is spent indoors on average, around the world[1]; C_F the conversion factor from absorbed dose in air to effective dose received by adults. A value of 0.7 Sv Gy^{-1} was used for the conversion factor. $D_{R\gamma}$ (nGy h^{-1}) was estimated using data and formula as follows[3]:

$$D_{R\gamma} = 0.12 \cdot A_{Ra} + 0.14 \cdot A_{Th} + 0.0096 \cdot A_K \quad (8)$$

where 0.12, 0.14 and 0.0096 nGy h^{-1} per Bq kg^{-1} are the conversion coefficients given only for superficial materials for ^{226}Ra, ^{232}Th and ^{40}K and A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively (in Bq kg^{-1}).

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Annual effective dose due to the indoor radon exposure

The annual effective dose (E_{Rn}, mSv) received by the inhabitants due to the indoor radon and its progeny was estimated by adopting the following model given in the United Nations Scientific Committee on the Effects of Atomic Radiation report[1]:

$$E_{Rn} = C_{Rn} \cdot F \cdot 8766 \text{ h y}^{-1} \cdot O_F \cdot D_C \cdot 10^{-6} \qquad (9)$$

where C_{Rn} is the indoor radon concentration (Bq m^{-3}), F the equilibrium equivalent concentration factor taken as 0.4 for inhabitants, O_F the indoor occupancy factor taken as 0.8 implying that 80 % of time is spent indoors and D_C the dose conversion factor taken as 9 nSv h^{-1} per Bq m^{-3}.

RESULTS AND DISCUSSION

The values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the granite samples together with the statistical uncertainty (1σ) are given in Table 1. From Table 1, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K varied from 10 to 187 Bq kg^{-1} with a mean of 80 ± 11 Bq kg^{-1}, 16–354 Bq kg^{-1} with a mean of 101 ± 17 Bq kg^{-1} and 104–1630 Bq kg^{-1} with a mean of 974 ± 102 Bq kg^{-1}, respectively. As shown the radionuclide contents in the granite samples show variations depending upon origin, geologic and geochemical characteristics of the granite samples. The mean values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are significantly (approximately two times) higher than the earth's crust mean values of 32, 45 and 420 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively[1]. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are comparable to those reported by other studies[15, 16, 21–26].

The EXR_S and the EXR_M were estimated for the model room (4 m × 5 m × 2.8 m) with walls and floor covered with a 3 cm thickness of granite tiles and a density of 2600 kg m^{-3} using Equations (3) and (4), respectively. It was assumed that η is equal to 0.45 because η ranged from <0.025 to 0.45[16]. The estimated values of the EXR_S and the EXR_M for the granite samples given in the third and fourth columns of Table 2 varied from 1.3 to 24.8 Bq m^{-2} h^{-1} with a mean of 10.5 ± 1.5 Bq m^{-2} h^{-1} and 0.03–0.64 with a mean of 0.27 ± 0.04 Bq kg^{-1} h^{-1}, respectively. As can be seen in Table 3, there are discrepancies between the values of EXR_S and the EXR_M reported and this levels estimated in the present study are comparable to those.

Table 1. The values of the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and gamma index (I_γ).

Sample no.	Commercial name	Activity concentration (Bq kg^{-1})			I_γ
		^{226}Ra	^{232}Th	^{40}K	
1	Bianco sardo	105 ± 4	16 ± 1	557 ± 42	0.6
2	Giresun vision	180 ± 10	176 ± 11	1550 ± 110	2.0
3	Bergama grey	70 ± 4	85 ± 5	841 ± 71	0.9
4	St. Gabriel	25 ± 1	30 ± 2	500 ± 43	0.4
5	Peacock green	15 ± 1	60 ± 3	1210 ± 105	0.8
6	Aksaray pasture	52 ± 2	58 ± 3	681 ± 56	0.7
7	Aksaray pink	50 ± 3	74 ± 3	938 ± 70	0.8
8	Baladan green	55 ± 4	61 ± 3	805 ± 61	0.8
9	Bandırma	187 ± 12	184 ± 12	1600 ± 131	2.1
10	Ayvalık grey	68 ± 5	86 ± 6	825 ± 71	0.9
11	Green butterfly	10 ± 1	77 ± 4	1630 ± 128	1.0
12	Tan brown	23 ± 1	123 ± 6	1205 ± 96	1.1
13	RP 302 Pink	123 ± 7	162 ± 7	1345 ± 121	1.7
14	CB 401 Red	120 ± 5	142 ± 6	1289 ± 119	1.5
15	CB Red	115 ± 5	124 ± 7	1281 ± 101	1.4
16	New imperial red	85 ± 5	354 ± 22	1325 ± 90	2.5
17	CS 202 Black	62 ± 4	61 ± 4	135 ± 11	0.6
18	Balmoral red	25 ± 2	16 ± 1	104 ± 10	0.2
19	Rosy pinky	90 ± 6	19 ± 1	562 ± 46	0.6
20	African red	132 ± 8	156 ± 10	1465 ± 117	1.7
21	Absolute black	32 ± 2	22 ± 1	223 ± 16	0.3
22	Red	127 ± 7	126 ± 8	1347 ± 97	1.5
	Min	10	16	104	0.2
	Max	187	354	1630	2.5
	Mean ± SE	80 ± 11	101 ± 17	974 ± 102	1.1 ± 0.1

[a]SE, standard error.

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Table 2. Estimated values of the EXR_S, EXR_M, indoor radon concentration (C_{Rn}), external annual effective dose (E_γ) and annual effective dose due to the indoor radon exposure (E_{Rn}) for the granite samples.

Sample no.	Commercial name	EXR_S (Bq m^{-2} h^{-1})	EXR_M (Bq kg^{-1} h^{-1})	C_{Rn} (Bq m^{-3})	E_γ (mSv)	E_{Rn} (mSv)
1	Bianco sardo	13.9	0.36	259	0.10	6.5
2	Giresun vision	23.9	0.61	444	0.30	11.2
3	Bergama grey	9.3	0.24	173	0.14	4.4
4	St. Gabriel	3.3	0.08	62	0.06	1.6
5	Peacock green	2.0	0.05	37	0.11	0.9
6	Aksaray pasture	6.9	0.18	129	0.10	3.3
7	Aksaray pink	6.9	0.18	129	0.13	3.3
8	Baladan green	7.2	0.19	134	0.11	3.4
9	Bandırma	24.8	0.64	461	0.31	11.6
10	Ayvalık grey	9.0	0.23	168	0.14	4.2
11	Green butterfly	1.3	0.03	23	0.14	0.6
12	Tan brown	3.0	0.08	56	0.15	1.4
13	RP 302 Pink	16.3	0.42	303	0.25	7.7
14	CB 401 Red	16.0	0.41	297	0.23	7.5
15	CB Red	15.3	0.39	284	0.21	7.2
16	New imperial red	11.3	0.29	210	0.36	5.3
17	CS 202 Black	8.2	0.21	152	0.08	3.8
18	Balmoral red	3.3	0.09	62	0.03	1.6
19	Rosy pinky	11.9	0.31	222	0.09	5.6
20	African red	17.5	0.45	325	0.25	8.2
21	Absolute black	4.2	0.11	78	0.04	2.0
22	Red	16.9	0.43	314	0.22	7.9
	Min	1.3	0.03	23	0.03	0.6
	Max	24.8	0.64	461	0.36	11.6
	Mean $\pm$ SE	10.6 ± 1.5	0.27 ± 0.04	196 ± 27	0.16 ± 0.02	5.0 ± 0.7

SE, standard error.

Table 3. Results of EXR_S and EXR_M of granite samples from the various countries with the present study.

Country	EXR_S (Bq m^{-2} h^{-1})	EXR_M (Bq kg^{-1} h^{-1})	References
Brasil	0.6–21.0	—	Anjos *et al.*[11]
Egypt	0.3–1.1	—	El-Dine *et al.*[27]
Italy	—	0.0043–0.25	Righi and Bruzzi[13]
Japan	—	1.04–1.97	Sakoda *et al.*[28]
Greece	Up to 3.5	Up to 0.24	Stoulos *et al.*[29]
Canada	0.3–0.5	—	Chen *et al.*[20]
India	<0.1–2.1	—	Sundar *et al.*[30]
Taiwan	0.1–1.0	—	Hassan *et al.*[31]
China	0.4–13.3	—	Hassan *et al.*[31]
Turkey	0.9	—	Topçu *et al.*[32]
Turkey	1.3–24.8	0.03–0.64	This study

The indoor radon concentration (C_{Rn}) was estimated using Equation (5). It was assumed that S/V equals 2 m^{-1} and λ_V equals 0.1 h^{-1} for poor ventilation. The calculated values of C_{Rn} for the granite samples given in the fifth column of Table 2 were found to be in the range of 23–461 Bq cm^{-3}, with a mean of 196 ± 27 Bq cm^{-3}. The design level for indoor radon exposure in future constructions set by the Turkish Standard TS 12614[33] corresponds to an annual mean radon gas concentration of 200 Bq m^{-3}, while the recommended value for remediation of indoor radon exposure in existing building set by the Turkish Radiation Safety Regulation is 400 Bq m^{-3}[34]. Considering upper levels recommended by the TS 12614 and Turkish Radiation Safety Regulation, it can be seen that 52 % of the granite samples show indoor radon concentrations <200 Bq m^{-3} and only two samples (Bandırma and Giresun vision) show values >400 Bq m^{-3}. If λ_V equals 0.5 h^{-1} for normal ventilation, the calculated values of C_{Rn} vary from 5 to 98 Bq cm^{-3} with a mean of 42 ± 6 Bq cm^{-3}.

The estimated values of the gamma index (I_γ) for the granite samples given in the last column of Table 1, except for Giresun vision, Bandırma and New imperial red samples are lower than the criterion of 2 corresponding to an effective dose 0.3 mSv.

The values of the external annual effective dose (E_γ) and annual effective dose due to the indoor radon exposure (E_{Rn}) estimated for the granite samples are given in the sixth and seventh column of

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Table 2, respectively. As can be seen from Table 2, the values of the E_γ and E_{Rn} ranged from 0.03 to 0.36 mSv and 0.6 with a mean of 0.16 ± 0.02 and 0.6–11.6 mSv with a mean of 5.0 ± 0.7 mSv, respectively. The values of the E_γ are lower than the exemption level of the annual effective dose of 0.3 mSv except for Giresun vision, Bandırma and New imperial red samples. The values of the E_{Rn} are below the recommended action level of 3–10 mSv by ICRP 65[35] except for Giresun vision and Bandırma samples.

CONCLUSIONS

This study presents the results of the measurement natural radioactivity in some commercially granite samples used as decorative or covering building materials in Turkey, and estimation of radon exhalation rates and indoor radon concentrations using indirect methods and assessment of the radiological hazards estimating the gamma index, external annual effective dose and annual effective dose due to the indoor radon exposure. As can be seen clearly from Tables 1–3, the majority of the obtained results are within the recommended safety limits. In view of the latest scientific data, the WHO proposes a reference level of 100 Bq m^{-3} to minimise health hazards due to indoor radon exposure. However, if this level cannot be reached under the prevailing country-specific conditions, the chosen reference level should not exceed 300 Bq m^{-3}, which represents $\sim$10 mSv y^{-1} according to recent calculations by the International Commission on Radiation Protection. But, the results show that the indoor radon concentrations ventilated adequately (by taking λ_V=0.5 h^{-1}) will be lower than the reference level of 100 Bq m^{-3} recommended by the WHO. Consequently, the granite samples examined in this study do not contribute significantly to increase the indoor radon concentration under normal ventilation conditions.

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