



Measurement of radioactivity and radon exhalation rate in different kinds of marbles and granites

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Abstract

Geological materials usually contaminated with naturally occurring radioactive materials (NORM) have become a focus of great attention. These NORM under certain conditions can reach hazardous contamination levels. Some contamination levels may be sufficiently severe that precautions must be taken.

The present study deals with 60 geological samples (marble and granite) from both Egyptian and foreign locations. The studied samples were analyzed and the concentrations in Bq/kg dry weight of radioisotopes were determined by gamma-ray spectrometry using hyper-pure germanium (HPGe) detector in Bq/kg dry weight. The absorbed dose rate due to the natural radioactivity in the samples under investigation ranged from 2.45 ± 0.07 to 64.44 ± 1.93 nGy/h for marble and from 41.55 ± 1.25 to 111.94 ± 3.36 nGy/h for granite. The radium equivalent activity varied from 5.46 ± 0.16 to 150.52 ± 4.52 Bq/kg for marble samples and from 229.52 ± 6.89 to 92.16 ± 2.76 Bq/kg for granite. The representative external hazard index values for the corresponding samples are also estimated and given.

The radon exhalation rates for marble and granite samples were also calculated by using solid state nuclear track detector (CR-39). The value of radium exhalation rate varied from 8.0 ± 2.39 to 30.20 ± 5.06 Bq/m²/d for marble and 6.89 ± 1.72 to 25.79 ± 4.38 Bq/m²/d for granite and the effective radium content was found to vary from 1.700 ± 0.51 to 6.42 ± 1.08 Bq/kg for marble and 1.29 ± 0.32 to 5.63 ± 0.96 Bq/kg for granite. The values of the radon exhalation rate and effective radium content are found to correspond with the values of uranium concentration measured by the HPGe detector in the corresponding sample. © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

Monitoring of any release of radioactivity to the environment is important for environmental protection. Rapid and accurate methods for the assay of radioactivity are essential. Low level gamma-ray spectrometry is suitable for both qualitative and quantitative determinations of gamma-ray emitting nuclides in the environment (IAEA, 1989).

The use of solid state nuclear track detectors (SSNTDS) is a convenient technique for low activity measurements since it is of low cost, is a simple

operation, has high registration sensitivity and has the possibility of use for long period exposures without any fading.

Most of the dose rate of natural radioactivity is due to the effect of primordial radionuclides, although the value take into account the contributions from cosmic radiation and cosmogenic radionuclides. In fact, only about 15% of the total effective dose equivalent derives from exposure to cosmic radiation, and about 0.6% is attributable to cosmogenic radionuclides. The members of the radioactive decay chains of ²³²Th (14%), ²³⁵U and ²³⁸U (55.8%), along with ⁴⁰K (13.8%) are responsible for the main contributions to the dose from natural radiation, while a mere 0.3% is due to the effect of ⁸⁷Rb (Luigi et al., 1997).

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Natural radioactivity concentration levels were also determined where this level contributed to the radiation dose received. Radioactivity in the environment consists of the three well-known radioactive series: the uranium series originates with ^{238}U , the thorium series originates with ^{232}Th and the actinium series originates with ^{235}U . As the ^{235}U to ^{238}U ratio is less than 1%, the contribution of ^{235}U to the environmental dose is very small. There are also several singly occurring radionuclides, the most important one is ^{40}K because it is a gamma-ray emitter in addition to beta decays and therefore contributes significantly to the gamma radiation exposure. Other pure beta particle emitters such as ^{87}Rb do not increase the radiation dose to man to any significant value, as beta particles are locally absorbed.

Emanation of radon (^{222}Rn) is associated with the presence of radium and its ultimate precursor uranium in the ground. The inhalation of its short-lived daughter products is a major contributor to the total radiation dose to exposed subjects (UNSCEAR, 1993). Due to the use of marbles and granites in building and decoration and the exposure of persons during a long time, this study is performed to measure the concentrations of the individual natural gamma-ray emitters in the uranium, thorium, actinium series and potassium in marble and granite samples. The radionuclides concentrations in the different samples are compared with the exempt levels proposed by the IAEA, which are of scientific interest to health physics and of economic interest to manufactures.

2. Experimental work

2.1. Sampling and sample preparation for γ -spectroscopy

Eight types of marble were collected from different locations, four are locally obtained: Type 1 (Galala) from Zafarana in Ghardaka, type 2 (Fleto) from El Hassana in Sinai, type 3 (Green marble) from Qena, type 4 (Assiout) from Assiout. The remaining marble types are imported: type 5 (black) from Spain, type 6 (White) from Greece, type 7 (Karara) from Italy and type 8 (Green) from India. Also there are four granite samples, the first type (9) is from East desert, the second type (10) is from Cleopatra Company, type (11) is from Pharana Company and type (12) is from Alpha Company. However, the last three types are artificial granite which are produced by the companies. Five samples are obtained from each type for accuracy.

Preparation of the collected samples for γ -ray measurements were carried out by drying the sample in an oven at a temperature of 110°C . The volume of the prepared samples were weighed. The samples were hermetically sealed in cylindrical plastic boxes with a volume of 350 cm^3 .

The samples were sealed tightly to limit the possible escape of radon. The samples were carefully sealed for 4

weeks to reach secular equilibrium between ^{226}Ra and ^{232}Th to be ready for γ -ray measurements using the HPGe detector and a multichannel analyzer with 8096 channel.

2.2. Experimental method for gamma spectroscopy

The gamma-ray system (Canberra coaxial hyper pure germanium detector) has a photo peak efficiency of 30% and energy resolution of 1.8 keV full-width at half-maximum (FWHM) for the 1332 keV gamma-ray line of ^{60}Co . A model 747 Canberra lead shield is used with the detector and has a 0.040 in tin and 0.062 in copper graded liner to prevent interference by lead X-rays. The calibration of the spectrometer was carried out by using standard point sources (^{60}Co , ^{226}Ra , ^{241}Am). Absolute efficiency calibration curves are calculated for activity determination of the sample by using standard ^{232}Th and ^{238}U with activities of 1333.96 and 2120.37 Bq, respectively, contained in the same cylindrical bottles as the samples. The standards and the samples were prepared with a uniform geometry. An empty bottle with the same geometry was measured for subtracting the background. The quality assurance of the measurements was carried out by a daily efficiency and energy calibration and repeating each sample measurement. The counting time was about 70,000 s to obtain the γ -spectrum with good statistics. The gamma-ray transitions of energies 351.9 keV (^{214}Pb), 609.3 keV, 1120.3 keV (^{214}Bi) and 1764 keV (^{214}Bi) were used to determine the concentration of the ^{238}U series. The gamma-ray transitions of energies 338.4 keV (^{228}Ac), 583.3 keV (^{208}Tl), 2614 keV (^{208}Tl) and 911.1 keV (^{228}Ac) were used to determine the concentration of the (^{232}Th) series. The 1460 keV gamma-ray transition ^{40}K was used to determine the concentration of ^{40}K in different samples.

2.3. Experimental method for radon measurement

The concentration and exhalation rate of radon can be made using CR-39 detectors because of their capability to register tracks at different levels of registration sensitivity. The calibration of CR-39 should be performed where the integrating radon's concentration is known (Khan and Varshney 1990). The CR-39 detectors used in this work were supplied by Pershore Mouldings, Ltd., UK, in the form of large sheets which were cut into $2\text{ cm} \times 2\text{ cm}$ squares. Each sample was placed in a glass cylinder of radius 3.5 cm and 10 cm length. Dosimeters were prepared by putting two CR-39 detectors in the bottom of the chamber cover. The cylindrical container was sealed; the samples were stored for at least 30 days.

The films were then exposed to radon and its daughters in the chamber for a known period of time. The exposure of the detectors, followed by etching of the

tracks left in the film provided the concentrations of radon and its daughters.

The optimum conditions for etching CR-39 (Mansy et al., 1998) were 6.25% N-NaOH at 70°C and 6h etching time while stirring the samples at 50 rev/min. After etching, the detector was washed in distilled water, dipped for a few seconds in 3% acetic acid solution, washed again and allowed to dry in air. Using an optical microscope at 400 × objective lens, the number of tracks in 50 fields were scanned for each detector to determine the track density per m³ (Khan et al., 1990).

3. Results and discussion

3.1. For gamma-ray spectroscopy

About 60 samples were investigated. About 17 resolved energy photo peaks were observed. Eight photo peaks were from the uranium series, eight were from the

thorium series and one was from ⁴⁰K. Figs. 1(a and b) show partial spectra of green Egyptian marble and the green Indian one. Fig. 2 shows a partial γ -ray spectrum of granite sample (9).

The average radionuclide activity concentration in Egyptian and foreign marble samples are reported in Tables 1 and 2, respectively. Also the activity concentrations for granite are reported in Table 3.

²³⁸U is the parent of the uranium series and is considered environmentally very important as it includes important radionuclides. Table 4 lists the average value of activities, the maximum value is in sample (1), 147.59 ± 4.73 Bq/kg and the minimum value is in sample (6), 3.10 ± 0.09 Bq/kg.

²³²Th is the parent of the thorium series. It is also very important from the environmental point of view, as it includes many essential radioisotopes. The maximum level was observed in sample (9), 63.10 ± 1.89 Bq/kg and the minimum level in sample (7), 1.16 ± 0.03 Bq/kg.

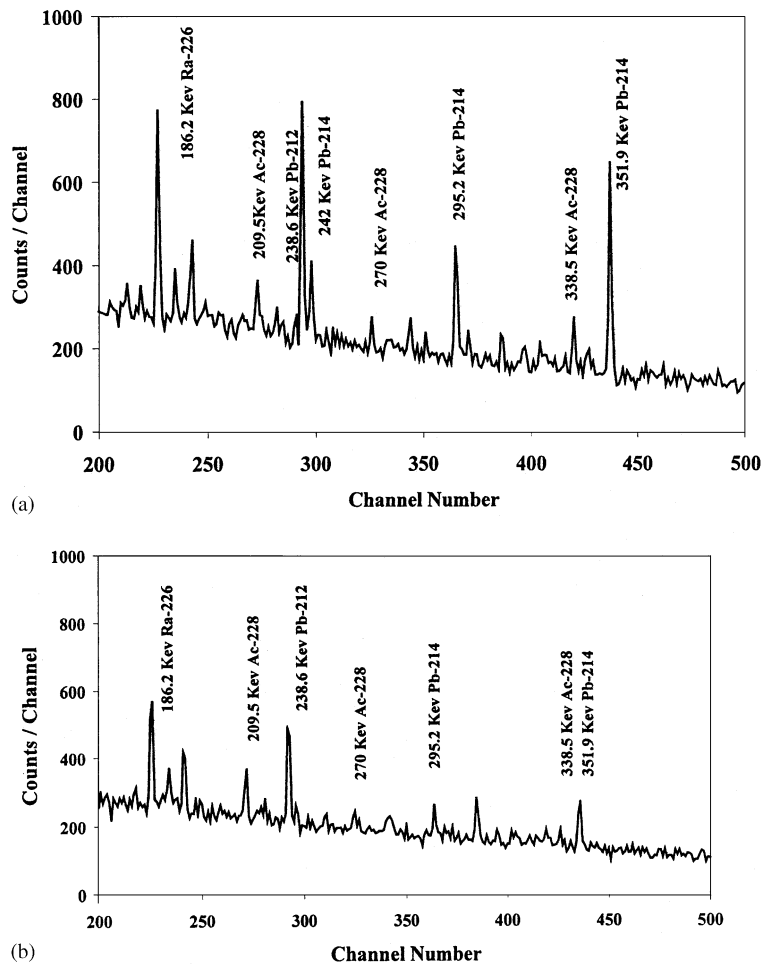


Fig. 1. (a) Partial spectrum for local Egyptian green marble sample. (b) Partial spectrum for the Indian green marble sample.

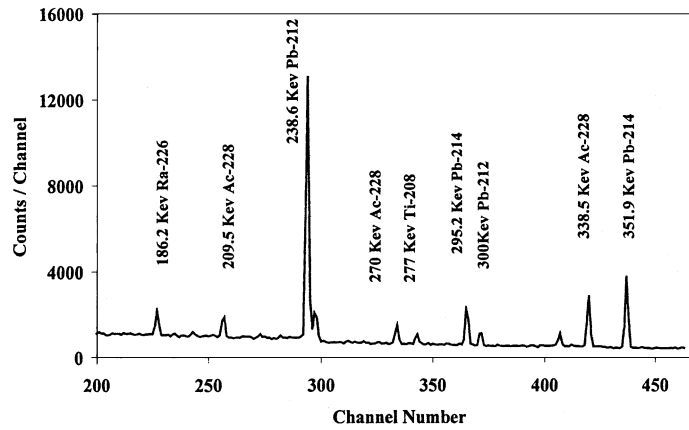


Fig. 2. Partial spectrum for granite sample (9).

Table 1
Activity concentration for different Egyptian marble samples (in Bq/kg)

Nuclide	Sample 1	Sample 2	Sample 3	Sample 4
U-series				
²²⁶ Ra	182.01 ± 5.46	23.63 ± 0.71	7.74 ± 0.23	4.92 ± 0.15
²¹⁴ Pb	147.40 ± 4.42	15.21 ± 0.46	4.39 ± 0.13	2.88 ± 0.09
²¹⁴ Bi	113.36 ± 3.40	13.29 ± 0.40	4.36 ± 0.13	4.80 ± 0.14
Th-series				
²²⁸ Ac	1.76 ± 0.05	L.L.D	2.79 ± 0.08	L.L.D
²¹² Pb	—	—	3.62 ± 0.11	1.59 ± 0.05
²⁰⁸ Tl	L.L.D	L.L.D	2.27 ± 0.07	1.47 ± 0.04
⁴⁰ K	5.79 ± 0.17	8.36 ± 0.25	43.35 ± 1.30	5.39 ± 0.16

Table 2
Activity concentration for different foreign marble samples (in Bq/kg)

Nuclide	Sample 5	Sample 6	Sample 7	Sample 8
U-series				
²²⁶ Ra	5.97 ± 0.17	5.86 ± 0.16	7.74 ± 0.23	6.10 ± 0.18
²¹⁴ Pb	2.91 ± 0.09	1.55 ± 0.05	1.55 ± 0.05	0.10 ± 0.03
²¹⁴ Bi	2.94 ± 0.09	1.89 ± 0.06	5.99 ± 0.18	3.04 ± 0.09
Th-series				
²²⁸ Ac	2.08 ± 0.06	1.37 ± 0.04	L.L.D	L.L.D
²¹² Pb	2.71 ± 0.08	1.53 ± 0.05	—	—
²⁰⁸ Tl	1.61 ± 0.05	1.32 ± 0.04	1.16 ± 0.03	1.21 ± 0.04
⁴⁰ K	32.41 ± 0.97	17.44 ± 0.52	13.45 ± 0.40	4.95 ± 0.15

The value of ⁴⁰K ranged from 4.95 ± 0.15 in sample (8) to 1335.18 ± 40.06 Bq/kg in sample (9). Comparing sample (3) and sample (8) we notice that the concentration of natural radioactivity in green Indian marble

is lower than the concentration in the Egyptian marble (sample 3) as shown in Figs. 1 (a and b). The activity concentrations for U, Th and K in marble samples are shown in Fig. 3. Also Fig. 4 shows

Table 3
Activity concentration for granite samples (in Bq/kg)

Nuclide	Sample 9	Sample 10	Sample 11	Sample 12
U-series				
²²⁶ Ra	30.23 ± 0.91	121.75 ± 3.65	83.99 ± 2.52	65.27 ± 1.96
²¹⁴ Pb	34.78 ± 1.04	135.51 ± 4.07	89.62 ± 2.69	70.67 ± 2.12
²¹⁴ Bi	24.66 ± 0.74	132.39 ± 3.97	86.57 ± 2.59	69.87 ± 2.09
Th-series				
²²⁸ Ac	49.05 ± 1.47	54.62 ± 1.64	62.16 ± 1.86	12.24 ± 0.37
²¹² Pb	65.39 ± 1.96	—	—	—
²¹² Bi	78.46 ± 2.35	62.38 ± 1.87	53.55 ± 1.61	8.10 ± 0.24
²⁰⁸ Tl	59.50 ± 1.78	57.08 ± 1.71	59.91 ± 1.79	10.43 ± 0.31
⁴⁰ K	1335.18 ± 40.0	238.06 ± 7.14	643.56 ± 19.30	127.10 ± 3.81

Table 4
Average activities (Bq/kg), dose rate (nGy/h), radium equivalent (Bq/kg), and the external hazard index for different granite and marble samples

Sample	²³⁸ U	²³² Th	⁴⁰ K	Dose rate (nGy/h)	Ra _{eq} (Bq/kg)	H _{ex}
1	147.59 ± 4.73	1.76 ± 0.05	5.79 ± 0.17	64.44 ± 1.93	150.52 ± 4.52	0.4
2	17.38 ± 0.52	L.L.D	8.36 ± 0.25	7.78 ± 0.23	17.97 ± 0.54	0.05
3	5.49 ± 0.16	2.89 ± 0.09	43.36 ± 1.30	6.11 ± 0.18	12.66 ± 0.38	0.04
4	4.20 ± 0.13	1.53 ± 0.05	5.39 ± 0.16	3.03 ± 0.09	6.77 ± 0.20	0.02
5	3.94 ± 0.12	2.13 ± 0.06	32.41 ± 0.97	4.48 ± 0.13	9.26 ± 0.28	0.03
6	3.10 ± 0.09	1.41 ± 0.04	17.44 ± 0.52	3.00 ± 0.09	6.34 ± 0.19	0.02
7	5.09 ± 0.15	1.16 ± 0.03	13.45 ± 0.40	3.52 ± 0.11	7.69 ± 0.23	0.02
8	3.38 ± 0.10	1.21 ± 0.04	4.95 ± 0.15	2.45 ± 0.07	5.46 ± 0.16	0.02
9	29.89 ± 0.80	63.10 ± 1.89	1335.18 ± 40.06	111.94 ± 3.36	213.58 ± 6.41	0.6
10	129.88 ± 3.89	58.03 ± 1.74	238.06 ± 7.14	104.12 ± 3.12	229.52 ± 6.89	0.85
11	86.73 ± 2.60	58.54 ± 1.76	643.56 ± 19.3	103.05 ± 3.09	170.44 ± 5.11	0.59
12	68.60 ± 2.06	10.26 ± 0.31	127.1 ± 3.81	41.55 ± 1.25	92.16 ± 2.76	0.25

the activity concentration for U, Th and K in granite samples.

Radium equivalent activity (Ra_{eq}). The distribution of ²²⁶Ra, ²³²Th and ⁴⁰K in building material is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of materials containing different amounts of ²²⁶Ra, ²³²Th and ⁴⁰K. It is calculated through the following relation (Ahmed, 1999; Tufail et al., 1992):

$$Ra_{eq} = (C_{Th} \times 1.43) + C_{Ra} + (C_K \times 0.077),$$

where C_{Th} is the ²³²Th activity concentration Bq/kg, C_{Ra} the ²²⁶Ra activity concentration Bq/kg and C_K the ⁴⁰K activity concentration Bq/kg.

The highest value of radium equivalent in marble is 150.52 ± 4.25 (Bq/kg) and in granite is 229.52 ± 6.89 Bq/kg. It is observed that the calculated radium equivalent is lower than the recommended maximum value 370 Bq/kg (OECD, 1979; Beretka and Mathew,

1985). It is assumed that 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of ²³²Th and 4810 Bq/kg of ⁴⁰K produce the same gamma-ray dose rate (Stranden, 1976).

External hazard index. The external hazard index due to the emitted gamma-rays of the samples are calculated and examined according to the following criterion:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1,$$

where A_{Ra}, A_{Th}, A_K were the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. The calculated average external hazard index was found to be less than unity.

The dose rate, the radium equivalent activity and the external hazard index for the samples under investigations are presented in Table 4.

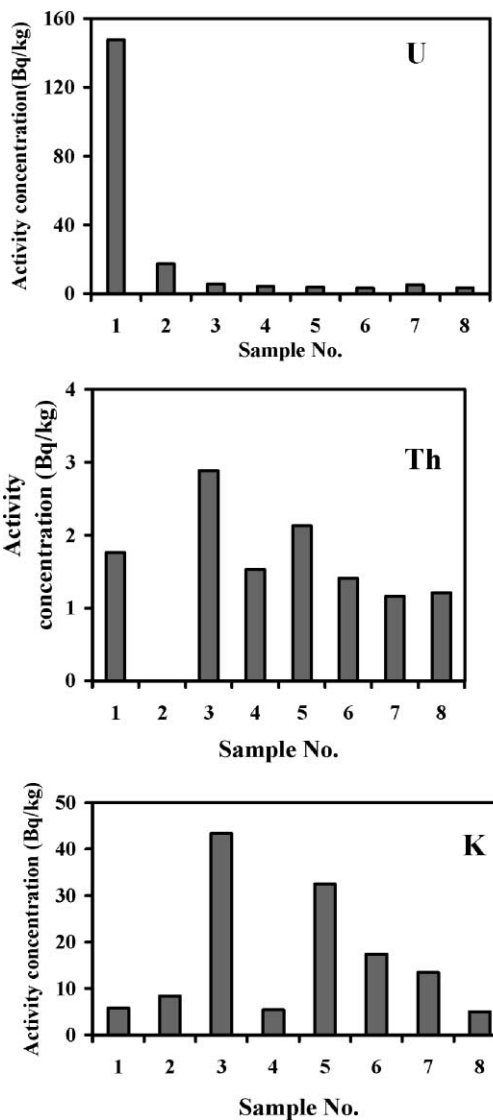


Fig. 3. Activity concentration for U, Th and K in marble samples.

3.2. For radon

Radon is an α -radioactive inert gas ($T_{1/2} = 3.825$ d), which can traverse through soil and other materials. The inhalation of Rn gas and its daughters represents a major health hazard. The exhalation rate of radon (Khan et al., 1990) can be calculated from the relation:

$$E_x = \frac{\rho V \lambda}{\xi A T_{\text{eff}}} \tag{1}$$

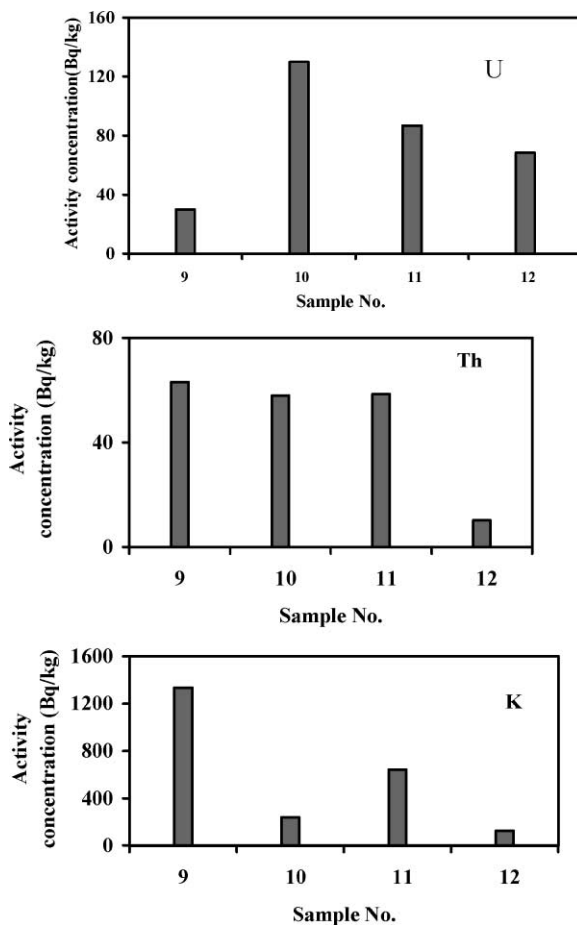


Fig. 4. Activity concentration for U, Th and K in granite samples.

The effective radium content is calculated from the equation:

$$C_R = \frac{\rho V}{\xi M T_{\text{eff}}} \tag{2}$$

where E_x is the radon exhalation rate ($\text{Bq}/\text{m}^2/\text{h}$), ρ the track density as measured by CR-39 detector, T/m^2 , V the effective volume of the cylindrical container (m^3), λ the radon decay constant, T_{eff} the effective exposure time $= (T + 1/\lambda(e^{-\lambda T} - 1))$, T the exposure time, A the area of the cylinder chamber, M the mass of the sample and ξ the detector efficiency.

Eight marble samples and four granite samples were collected for radioactive isotope determination. In order to evaluate the radon risk in a given atmosphere, it is necessary to check the relation between the activity concentrations of uranium and radon for the samples under investigations. In the present work, we have

Table 5
The density, effective radium content, and exhalation rate of radon

Sample	Track density (T/m ²) × 10 ³	Eff. radium content (Bq/kg)	Exhalation rate (Bq/m ² /d)
1	7.55 ± 1.26	6.42 ± 1.08	30.20 ± 5.06
2	3.39 ± 0.58	2.89 ± 0.49	13.58 ± 2.31
3	2.76 ± 0.57	2.35 ± 0.49	10.36 ± 2.28
4	2.44 ± 0.62	2.07 ± 0.53	9.75 ± 2.49
5	2.00 ± 0.59	1.70 ± 0.51	8.00 ± 2.39
6	2.10 ± 0.52	1.79 ± 0.44	8.42 ± 2.09
7	3.07 ± 0.63	2.61 ± 0.54	10.28 ± 2.52
8	2.21 ± 0.58	1.88 ± 0.49	8.83 ± 2.31
9	1.53 ± 0.38	1.29 ± 0.32	6.89 ± 1.72
10	6.64 ± 1.13	5.63 ± 0.96	25.79 ± 4.38
11	4.44 ± 0.75	3.76 ± 0.64	17.65 ± 3.00
12	3.51 ± 0.74	2.97 ± 0.62	14.21 ± 2.98

applied solid state nuclear track detectors, (SSNTDS) CR-39, for the exhalation rate measurements.

It was assumed that the radionuclides were in equilibrium, i.e., the activity of each daughter was equal to the initial isotope of the series. The values of the radon exhalation rate and the effective radium content from the samples are found to correspond with the values of uranium concentrations measured by the hyper pure germanium detector in the corresponding sample. Results of the present work are summarized in Table 5. The radon exhalation rate for marble samples were found to vary from 8.00 ± 2.39 to 30.20 ± 5.06 Bq/m²/d. The effective radium content varied from 1.700 ± 0.51 to 6.42 ± 1.08 Bq/kg, the large variation may be attributed to the variation of uranium concentrations according to different kinds of marble. With respect to granite the radon exhalation rate varies from 6.89 ± 1.72 to 25.79 ± 4.38 Bq/m²/d and the effective radium content varied from 1.29 ± 0.32 to 5.63 ± 0.96 Bq/kg.

Comparing the global value 30 Bq/kg (UNSCEAR, 1993) with the present work, it is clear that, except for one sample, radon concentrations are much smaller than the global average. From Figs. 5–8 it is evident that the relation between the activity concentrations of uranium and radon exhalation rate, and the effective radium content for marble and granite is a linear one.

4. Conclusion

Environmental monitoring should be carried out for marbles and granites where people might be exposed to radioactivity. A combination between gamma-ray spectroscopy and solid state nuclear track detectors is recommended to determine radon and uranium con-

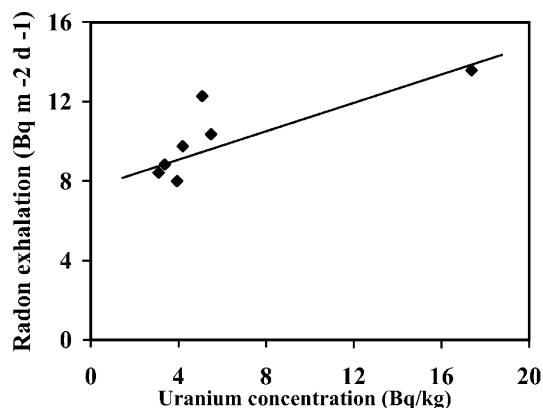


Fig. 5. Radon exhalation versus uranium concentration for marble samples.

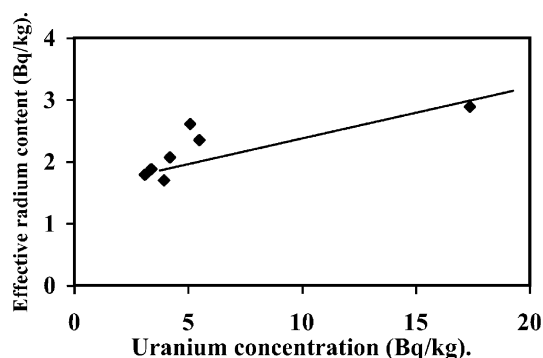


Fig. 6. Effective radium content versus U concentration for marble samples.

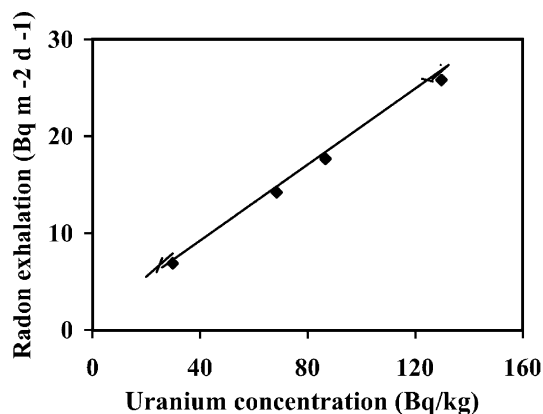


Fig. 7. Radon exhalation versus U for granite samples.

centrations. The levels of natural radioactivity in marble and granite samples were determined.

The studied marble samples can be classified according to their levels of radionuclides concentrations. In the case of uranium the highest level was found in the

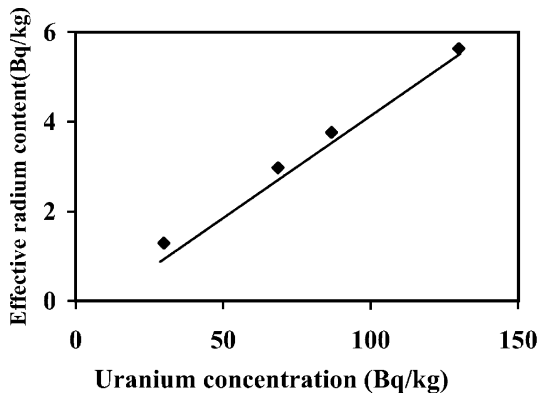


Fig. 8. Effective radium content versus U concentration for granite samples.

Galala marble sample and the lowest level in the white marble sample (from Greece). For thorium the highest level was found in Green marble (from Qena) and the lowest level in Karaka marble sample (from Italy), while for potassium the highest level was found in Green marble (from Qena) and the lowest level in the Green marble sample from India. Similarly, in granite samples, the highest level for uranium was found in the sample from Cleopatra Company and the lowest level in the granite sample from East desert, while with respect to thorium and potassium the highest levels was found in the sample from East desert and the lowest levels from Alpha Company. A comparison between the local and foreign samples was made. The results of this comparison showed that the local samples contain somewhat higher levels of radioactivity than that of the imported ones. Based on the results obtained, it is concluded that the radioactivity levels of marbles and granites are within the international recommended values (UNSCEAR, 1993). The relation between the activity

concentrations of uranium and radon exhalation rate is a linear one, so the knowledge of uranium concentrations gives a good estimate of the radon concentrations of the samples and its escape to the atmosphere.

According to the recommended values and the calculated external hazard index values the samples are acceptable for use as building materials and decoration.

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