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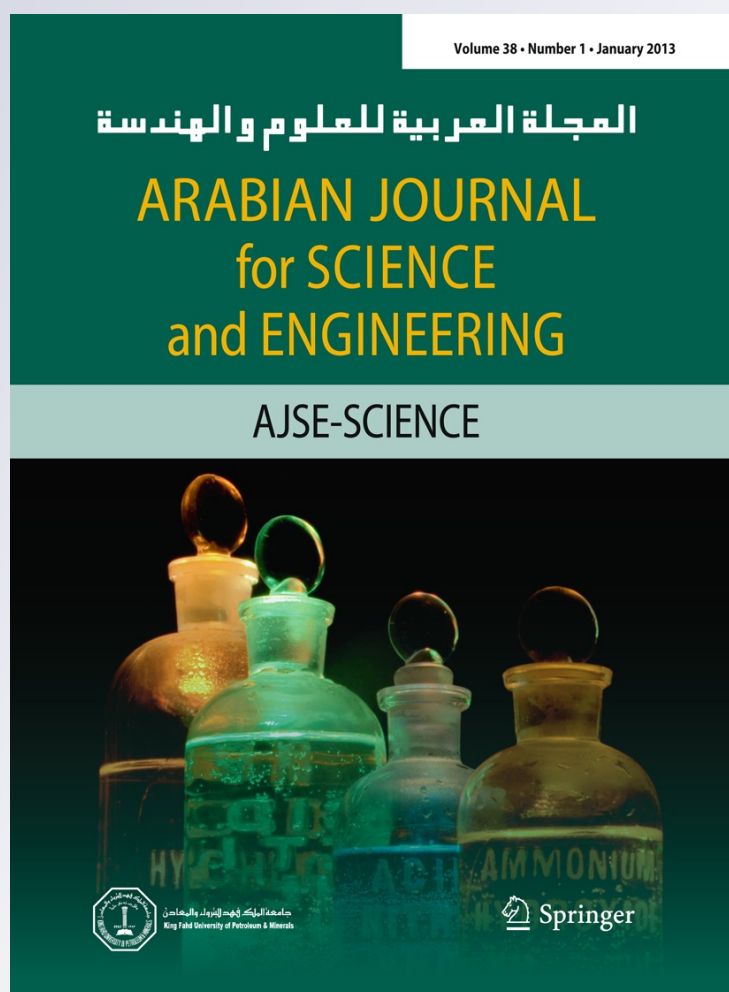
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# Measurement of Natural Radioactivity and Radon Exhalation Rate in Granite Samples Used in Palestinian Buildings

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**Abstract** The activity concentrations and radon exhalation rates of natural radionuclides were measured for 40 samples of different imported fabricated granite types used in constructing Palestinian dwellings to estimate the radiation exposure in the atmosphere using high-resolution gamma-ray spectrometry. The average activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides were found to vary from 18.6 to 151.2, 28.8 to 211.0 and 400.0 to 1,256.8  $\text{Bq kg}^{-1}$ , respectively. The overall average activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were 71.0, 82.0 and 780.8  $\text{Bq kg}^{-1}$ . The average radium equivalents were found to range from 100.5 to 517.6  $\text{Bq kg}^{-1}$  with a total average value of 248.2  $\text{Bq kg}^{-1}$ . The average absorbed dose rate was estimated at 1 m above ground level and a value of 118.2  $\text{nGy h}^{-1}$  is obtained. The measured average annual effective dose rates varied from 0.24 to 1.2  $\text{mSv year}^{-1}$ , with a total average value of 0.58  $\text{mSv year}^{-1}$ . The external hazard index measured average values were found to be within 0.27–1.4  $\text{Bq kg}^{-1}$  with a total average value of 0.67  $\text{Bq kg}^{-1}$ . The radioactivity concentration level estimated values varied from 0.7 to 3.7  $\text{Bq kg}^{-1}$  with a total average value of 1.8  $\text{Bq kg}^{-1}$ . The radon exhalation rate values range between 3.9 and 30.6  $\text{Bqm}^{-1} \text{ day}^{-1}$ . The effective radium content found to vary from 0.81 to 6.12 with an overall average value of 3.1  $\text{Bq kg}^{-1}$ .

**Keywords**  $^{226}\text{Ra}$  ·  $^{232}\text{Th}$  and  $^{40}\text{K}$  estimation in granite · Radon ·  $^{222}\text{Rn}$  exhalation rates · External hazard index

## الخلاصة

لقد تم قياس تراكيز النشاط الإشعاعي الطبيعي، ومعدل غاز الرادون المنطلق من نويدات ذات نشاط إشعاعي طبيعي لأربعين عينة مختلفة من الجرانيت المستورد والمستخدم في بناء المباني الفلسطينية، لقياس مستويات التعرض للنشاط الإشعاعي الطبيعي في الجو المحيط باستخدام مطيافية أشعة جاما ذات قدرة فصل عالية. وقد وجد أن متوسط التراكيز للنويدات  $^{40}\text{K}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$  تتراوح ما بين 18.6 – 151.2 بيكريل/كغم و 28.8 – 211.0 بيكريل/كغم و 400 – 1256.8 بيكريل/كغم، ومتوسط كلي مقداره 71.0 و 82.0 و 780.8 بيكريل/كغم، على التتابع.

لقد وجد أن متوسط نشاط الراديوم المكافئ يتراوح ما بين 100.5 – 517.6 بيكريل/كغم بمتوسط كلي مقداره 248.2 بيكريل/كغم<sup>1</sup>. ووجد أن متوسط معدل الجرعة الممتصة في العينات على ارتفاع متر واحد فوق مستوى سطح الأرض 118.2 نانوجراي/ساعة. أما المتوسط السنوي لمعدل الجرعة الفعالة فوجد أنه يتراوح بين 0.24 – 1.2 ملي سيفرت/سنة ومتوسط كلي مقداره 0.58 ملي سيفرت/سنة. أيضا تم تقدير مؤشرات الخطر الخارجي، ووجد أن قيمته تتراوح ما بين 0.27 و 1.4 بيكريل/كغم، ومتوسط كلي مقداره 0.67 بيكريل/كغم<sup>1</sup>. أما قيم مستويات تراكيز النشاط الإشعاعي المقدرة، فتتراوح ما بين 0.7 إلى 3.7 بيكريل/كغم<sup>1</sup> بمتوسط كلي 1.8 بيكريل/كغم<sup>1</sup>. أما معدل قيم غاز الرادون المنطلق فوجد أنها تتراوح ما بين 3.9 إلى 30.6 بيكريل/يوم. متر. وبالنسبة لمعدل الراديوم الفعال، فإن قيمة تتراوح بين 0.81 إلى 6.12 بمتوسط كلي 3.1 بيكريل/كغم<sup>1</sup>.

## 1 Introduction

Radioactive materials, such as potassium-40 ( $^{40}\text{K}$ ), uranium-238 ( $^{238}\text{U}$ ) and thorium-232 ( $^{232}\text{Th}$ ), are naturally presented in the earth's crust (soil, rock, water, and air) at different abundance levels [1,2]. Granite is a form of igneous rock, composed primarily of Quartz, Alkalie, and Feldspar, which contains some radionuclides [2]. For example, the white or pink feldspars contain  $^{40}\text{K}$ , the black biotites and the hornblendes contain  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ , and the zircon, apatite, sphene, minerals contain  $^{238}\text{U}$  and  $^{232}\text{Th}$  [3]. The presence of radium and its ultimate precursor uranium in the ground [2],

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is generally associated with emission of radon-222 ( $^{222}\text{Rn}$ ) from earth's crust [4–6].

Natural and the fabricated granite stones are both widely used worldwide as building materials, especially in porcelain fixtures, decoration, ornamental and monumental materials due to its elegant look and it is ease to clean. Internal and external radiation exposure from building materials creates prolonged exposure either by emitting gamma ( $\gamma$ ) radiations, or by releasing  $^{222}\text{Rn}$  gas [5].

The  $\gamma$ -radiation emitted during the decay of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  radioactive nuclides and their progenies, increase the whole body absorbed doses. Beta radiations ( $\beta$ -radiation) can enhance absorbed skin doses. The assigned worldwide average indoor effective dose due to  $\gamma$ -rays from building materials is estimated to be  $0.4 \text{ mSv year}^{-1}$  [2]. The level index,  $I_\gamma$ , is generally used to assess the  $\gamma$ -radiation hazard associated with the natural radionuclide in granite samples. In general, values of  $I_\gamma \leq 1$ , correspond to  $0.3 \text{ mSv year}^{-1}$ , while values  $< 3$  correspond to  $1 \text{ mSv year}^{-1}$ . Since, the highest dose rate recommended for population by international organizations and research groups is  $1 \text{ mSv year}^{-1}$ , all materials having  $I_\gamma \leq 3$  and  $I_\gamma > 3$  should be avoided [2–4].

The radium equivalent concentration index,  $\text{Ra}_{\text{eq}}$ , was introduced to compare specific activities of materials containing different amounts of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides. To keep the  $\gamma$ -radiation doses received from building materials below  $1.5 \text{ mSv year}^{-1}$ , one has to make sure that the maximum value of  $\text{Ra}_{\text{eq}}$  in building materials is  $< 370 \text{ Bq kg}^{-1}$ , and the external hazard index,  $H_{\text{ex}}$ , is also less than unity ( $H_{\text{ex}} < 1$ ). In such cases, the radiation hazard can be maintained as low as possible.

The second prolonged exposure is due to  $^{222}\text{Rn}$  and its radioactive daughters [6,7]. Due to its long half-life (3.8 days), outdoor as well as indoor  $^{222}\text{Rn}$  gas diffusion might occur from the earth's crust or from the buildings walls and floors. The indoor  $^{222}\text{Rn}$  exposure risk is higher in buildings with poor ventilation systems, where  $^{222}\text{Rn}$  gas accumulation resulted in higher indoor  $^{222}\text{Rn}$  concentrations [6]. The inhalation of  $^{222}\text{Rn}$  short-lived progeny is of a major contributor to the total radiation dose of exposed subjects [4]. In general,  $^{222}\text{Rn}$  contribution is about 55 % of the total dose received by the cells of the respiratory tract system [2–8]. Thus, the produced radioactive progeny of  $^{222}\text{Rn}$  in the human respiratory was deposited in the lung cells; hence the possibility of having lung cancer is increased [2,6,7]. The assigned USA average indoor concentration level is  $150 \text{ Bq m}^{-3}$  [2]. To characterize the building materials as an indoor radon source, assessment of the radon exhalation rate,  $E_x$ , from such materials is of great importance. The assessed  $E_x$  values can be used as indicators for the presence of uranium in granite samples.

Tremendous investigations and studies have been conducted worldwide on mapping natural radioactivity and  $E_x$

from building materials [9–21]. In this study, the activity concentrations of natural radionuclide and  $E_x$  measurements of different fabricated imported granite types used as construction materials in Palestinian buildings will be measured and compared to the assigned international levels.

## 2 Experiments

### 2.1 Sample Preparation

A total of 40 fabricated imported granite samples from Italy, India, South Africa, Brazil and Zimbabwe that are widely used as building and ornamental materials in West Bank, Palestine were collected from several commercial companies, construction sites and local suppliers. The average sample dimensions were around  $300 \text{ mm} \times 200 \text{ mm} \times 20 \text{ mm}$  and their masses vary from 2.8 to 3.6 kg. General information and brief descriptions of the investigated samples are exhibited in Table 1.

After the samples were collected, they are crushed into a fine powder type from which fine particle samples were obtained using a sieve of  $200 \mu\text{m}$  mesh size. Before performing measurements, samples were oven dried at  $110^\circ\text{C}$  for 8 h and then packed in 1 L Marenilli beaker and sealed for 4 weeks to reach secular equilibrium between  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  with their decay products.

### 2.2 Gamma Rays Spectroscopy

The radionuclide activity concentrations in the prepared samples were measured using an n-type coaxial lead shielded intrinsic Ortec high-purity germanium (HPGe) detector. The detector has an efficiency of about 15 %, energy resolution of 1.85 keV, full width to the half maximum (FWHM) for the 1332.5 KeV gamma line of  $^{60}\text{Co}$  and MCA with 8,000 channels [22]. An empty Marenilli beaker was used for the same period of time to assess the background concentrations of the  $\gamma$ -rays. To reduce the  $\gamma$ -rays background from building and cosmic rays, a cylindrical lead shield of 100 mm thickness is used to shield the detector from the surroundings environment. This shield is composed of three inner concentric shells of lead, cadmium and copper.

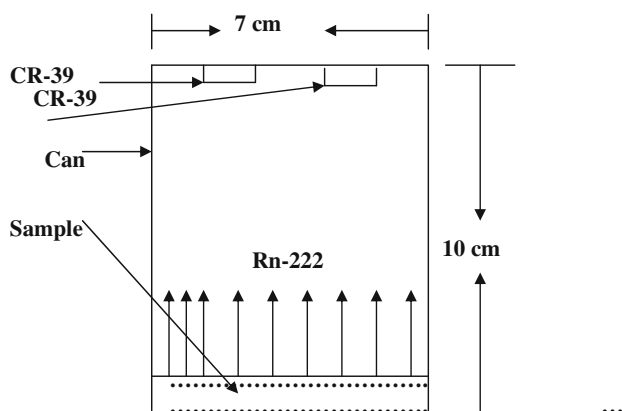
### 2.3 Spectroscopic Analysis

The quality assurance of the measurements was performed daily for energy calibration and activity measurements. A counting time of 70,000 s was used to obtain good measurements for  $\gamma$ -rays spectrum. The  $\gamma$ -rays transitions having energies of 351.9 keV ( $^{214}\text{Pb}$ ), 609.3 keV, 1,120.3 keV ( $^{214}\text{Bi}$ ) and 1,764 keV ( $^{214}\text{Bi}$ ) were used to determine the concentration of the  $^{238}\text{U}$  series. However, the  $\gamma$ -rays



**Table 1** Code number, commercial names, origins, density and colors of imported granite samples used in Palestinian buildings

Sample code	Commercial name	No. of samples	Country of origin	Density ( $\times 10^3 \text{ kg m}^{-3}$ )	Color
IGS <sub>1</sub>	Lampra	3	Italy	2.4	A mixture of red and black
IGS <sub>2</sub>	Rosa beta	3	Italy	2.5	A mixture of blue, red
IGS <sub>3</sub>	Galaxy	3	Italy	3.0	Black gold
IGS <sub>4</sub>	Barginza	3	Italy	2.5	Pink
IGS <sub>5</sub>	Ajami 1	2	Italy	2.7	Green light
IGS <sub>6</sub>	Tiger	3	Italy	2.9	Black
InGS <sub>7</sub>	Paradiso	3	India	2.6	Dark brown
InGS <sub>8</sub>	Sara	3	India	2.7	Light brown
SGS <sub>9</sub>	Seizerbaz	3	South Africa	2.8	A mixture of blue and black
SGS <sub>10</sub>	Oreiz	3	South Africa	3.0	Grey
SGS <sub>11</sub>	Nier-Africa	3	South Africa	2.9	Black
BGS <sub>12</sub>	Seizer-bright	2	Brazil	2.6	A mixture of blue and black
BGS <sub>13</sub>	Ajami 2	3	Brazil	2.7	Grey
ZGS <sub>14</sub>	Apsselto	3	Zimbabwe	3.0	Black



**Fig. 1** Schematic diagram of the sealed-can technique for measuring radon exhaled from a radium-containing material by means of a CR-39 alpha-particle sensitive track detector

transitions of energies 338.4 keV (<sup>228</sup>Ac), 583.3 keV (<sup>208</sup>Tl), 2,614 keV (<sup>208</sup>Tl) and 911.1 keV (<sup>228</sup>Ac) were used to determine the concentration of the <sup>232</sup>Th series. The 1,460 keV  $\gamma$ -rays transition was used to determine the concentration of <sup>40</sup>K in different samples.

#### 2.4 Radon Concentration and Radon Exhalation Rate

The concentration levels and exhalation rate of <sup>222</sup>Rn were measured using CR-39 detectors supplied by Pershore Mouldings, Ltd., UK, in the form of large sheets from which detectors of dimensions 10 mm  $\times$  10 mm were produced. Dosimeters were prepared by fixing two CR-39 detectors to the top of the inverted chamber cover of a glass cylinder of radius 35 mm and a length of 100 mm (see Fig. 1). The CR-39 calibrations have been made somewhere else [14].

The granite samples were stored in Marenilli beaker for at least 30 days to establish equilibrium between radium and radon before starting each measurement. The samples were

placed in a sealed dosimeter and the CR-39 detectors were then exposed to radon and its progeny in the chamber for 8 weeks after. After the exposure time, the detectors were collected and prepared for etching. The optimum conditions for CR-39 etching were: 6.25 N solution of NaOH at a temperature of 70°C and 6 h etching time to enhance the damaged tracks [23,24]. After etching, the detector was washed in distilled water, dipped for a few seconds in 3 % acetic acid solution, washed again and dried in hot air. Using an optical microscope at 150 $\times$  objective lens, the number of tracks in 30 fields was scanned for each detector to determine the track density per m<sup>3</sup> [14]. The tracks were counted and the concentrations of both radon and its progeny were determined according to Hafez et al. [23].

### 3 Calculations

#### 3.1 The Activity Concentrations

The activity concentration, *A*, values in the investigated samples were calculated using the following equation [25]:

$$A = \frac{(\text{cps})_{\text{net}}}{I \times \varepsilon_{\text{ff}} \times M}, \quad (\text{Bq kg}^{-1}) \quad (1)$$

where (cps)<sub>net</sub> is the number of counts per second [(cps)<sub>sample</sub> – (cps)<sub>background</sub>], and *I* is the intensity of the  $\gamma$ -line in a radionuclide [26]. Moreover,  $\varepsilon_{\text{ff}}$  is used to represent the measured efficiency for each observed  $\gamma$ -line and *M* is the mass of the sample in kilograms [26].

#### 3.2 The Radium Equivalent

The radium equivalent index, Ra<sub>eq</sub>, is generally introduced as the weighted sum of <sup>226</sup>Ra (daughter of <sup>238</sup>U), <sup>232</sup>Th and <sup>40</sup>K activities based on the assumption that 10 Bq kg<sup>-1</sup> of



$^{226}\text{Ra}$ ,  $7 \text{ Bq kg}^{-1}$  of  $^{232}\text{Th}$  and  $130 \text{ Bq kg}^{-1}$  of  $^{40}\text{K}$  will produce the same dose rates of  $\gamma$ -rays. Values of  $\text{Ra}_{\text{eq}}$  are calculated using the equation

$$\text{Ra}_{\text{eq}} = C_{\text{Ra}} + (C_{\text{Th}} \times 1.43) + (C_{\text{K}} \times 0.077), \quad (\text{Bq kg}^{-1}) \quad (2)$$

where  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively.

### 3.3 Absorbed Gamma Dose

The absorbed gamma dose,  $D_\gamma$ , values for the uniform distribution of radionuclides ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ ), 1 m above the ground surface, were calculated on the basis of guidelines adopted by UNSCEAR [2,4]. The conversion factors used to compute  $D_\gamma$  in air per unit activity concentration ( $\text{Bq kg}^{-1}$ ) in soil are  $0.462 \text{ nGy h}^{-1}$  for  $^{226}\text{Ra}$ ,  $0.621 \text{ nGy h}^{-1}$  for  $^{232}\text{Th}$ , and  $0.0417 \text{ nGy h}^{-1}$  for  $^{40}\text{K}$ . The values of  $D_\gamma$  were calculated by making use of the following equation [27]:

$$D_\gamma = 0.462C_{\text{Ra}} + 0.621C_{\text{Th}} + 0.0417C_{\text{K}}, \quad (\text{nGy h}^{-1}) \quad (3)$$

### 3.4 External Hazard Index

The external hazard index,  $H_{\text{ex}}$  values were calculated using Eq. (4) [26]. Besides, activity concentrations of 370, 259, and  $4,810 \text{ Bq kg}^{-1}$  used for  $^{226}\text{Ra}$  of  $^{226}\text{Ra}$   $^{40}\text{K}$ , respectively, were assumed to produce the same  $D_\gamma$  [26]. Accordingly, values of  $H_{\text{ex}}$  were calculated according to the equation [26]:

$$H_{\text{ex}} = \frac{C_{\text{Ra}}}{370} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4,810} \leq 1 \quad (4)$$

According to the Eq. (4),  $H_{\text{ex}}$ , should be less than unity ( $H_{\text{ex}} < 1$ ) to ensure negligible radiation hazard.

### 3.5 Annual Effective Dose Rates

The annual effective dose rate values, EDR, were calculated using a conversion coefficient of  $0.7 \text{ SvG year}^{-1}$  for converting absorbed dose in air into effective dose and assuming an indoor occupancy factor of 0.8 [2]. The indoor EDR in units of  $\text{mSv year}^{-1}$  was calculated according the following formula [26]:

$$\text{EDR} = \left[ D_r (\text{mGy h}^{-1}) \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \right] \times 10^{-6}, \quad (\text{mSv year}^{-1}) \quad (5)$$

### 3.6 Radioactivity Level Index, $I_\gamma$

The  $\gamma$ -radiation hazardous levels associated with the natural radionuclides in the granite samples were assessed by means

of radioactivity level index,  $I_\gamma$ . According to the European Commission guidelines, the representative level of  $I_\gamma$  values was estimated according to the following equation [26]:

$$I_\gamma = \frac{C_{\text{Ra}}}{150} + \frac{C_{\text{Th}}}{100} + \frac{C_{\text{K}}}{1500} \quad (6)$$

The index  $I_\gamma$  was correlated to the annual dose due to the excess external  $\gamma$ -radiation caused by superficial material.

### 3.7 Measurement of Radon Exhalation Rate

The radon exhalation rate of radon,  $E_x$ , can be calculated from the empirical relation [28]:

$$E_x = \frac{\rho V \lambda}{\xi A T_{\text{eff}}}, \quad (\text{Bq m}^{-2} \text{ h}^{-1}) \quad (7)$$

where  $\rho$  is the track density ( $\text{T/m}^2$ ) measured by CR-39 detector and  $V$  is the effective volume of the cylindrical container ( $\text{m}^3$ ). The radon decay constant,  $\lambda$ , has a value of  $7.57 \times 10^{-3} \text{ h}^{-1}$ , and the effective exposure time,  $T_{\text{eff}}$ , can be expressed as:  $T_{\text{eff}} = T + 1/\lambda(e^{-\lambda T} - 1)$ . The exposure time,  $T$ , is measured in seconds (s), and the area of the cylindrical chamber,  $A$ , is measured in  $\text{m}^2$ . The detector efficiency,  $\xi$ , has a value of 0.4 tracks/particle.

### 3.8 The Effective Radium Content, $C_R$

The effective radium content,  $C_R$ , is defined as radon exhalation rate per radon decay constant,  $\lambda$ .  $C_R$  is calculated according to the equation [28]:

$$C_R = \frac{E_x}{\lambda} = \frac{\rho V}{\xi M T_{\text{eff}}}, \quad (\text{Bq kg}^{-1}) \quad (8)$$

where  $M$  is the mass of the sample in kilograms.

## 4 Results and Discussion

### 4.1 Activity Concentrations

The measured average activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (in  $\text{Bq kg}^{-1}$ ) radionuclides in the investigated 40 granite samples are presented in Table 2.

As it can be seen from Table 2, the variations of the average activity concentration values of radionuclides in the investigated samples were as follows: from 18.6 to  $151.2 \text{ Bq kg}^{-1}$  with a total average value of  $71.0 \text{ Bq kg}^{-1}$  for  $^{226}\text{Ra}$ ; from 28.8 to  $211.0 \text{ Bq kg}^{-1}$  with an a total average value of  $82.0 \text{ Bq kg}^{-1}$  for  $^{232}\text{Th}$ ; and from 400.0 to  $1256.8 \text{ Bq kg}^{-1}$  with an a total average value of  $780.8 \text{ Bq kg}^{-1}$  for  $^{40}\text{K}$ . Besides, the obtained average activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were found to vary with the production

**Table 2** The average activity concentrations of radionuclides in imported granite samples used in Palestinian regions

Sample code	No. of sample	Activity concentrations (Bq kg <sup>-1</sup> )		
		<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
IGS <sub>1</sub>	3	118.0	110.2	1185.4
IGS <sub>2</sub>	3	151.2	189.0	1253.0
IGS <sub>3</sub>	3	86.5	73.5	600.0
IGS <sub>4</sub>	3	139.7	140.8	1256.8
IGS <sub>5</sub>	2	49.1	61.8	940.0
IGS <sub>6</sub>	3	41.1	39.6	834.0
InGS <sub>7</sub>	3	65.3	211.0	897.4
InGS <sub>8</sub>	3	52.0	63.5	1020.6
SGS <sub>9</sub>	3	57.6	67.2	862.2
SGS <sub>10</sub>	3	18.6	35.5	459.2
SGS <sub>11</sub>	3	46.6	28.8	425.0
BGS <sub>12</sub>	2	106.2	53.6	837.0
BGS <sub>13</sub>	3	28.0	29.2	400.0
ZGS <sub>14</sub>	3	35.1	43.9	451.3
	Total average	71.0	82.0	780.8

site of the granite and various colors. Moreover, the average activity concentrations for most samples were inversely proportional to the sample density. Accordingly, the average values of all activity concentrations in red and pink imported granites are found to be higher than those in black and gray granites, in general. The high concentration values of <sup>226</sup>Ra and <sup>232</sup>Th in granite samples were attributed to the presence of uranium mineralization and the construction materials of the rock as in K-feldspars, hematite, garnet and chlorites, and due to the presence of active faults and lineaments in the rocks.

The obtained results of the average activity concentrations for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K, for most samples, were higher than the corresponding assigned international radioactivity levels of 50, 50 and 500 Bq kg<sup>-1</sup>, respectively [4]. Specifically, concentration activities for <sup>226</sup>Ra and <sup>232</sup>Th were found to be 60 % higher than the world average; while <sup>40</sup>K concentration activity was found to be 70 % higher than the world average.

#### 4.2 Radiological Effects

The average values of C<sub>Ra</sub>, C<sub>Th</sub> and C<sub>K</sub> were employed to determine Ra<sub>eq</sub>, D<sub>γ</sub>, EDR, H<sub>ex</sub>, and I<sub>γ</sub>, for all investigated granite samples using Eqs. (2)–(6). The obtained results are summarized in Table 3.

As it can be seen from Table 3, the average values of Ra<sub>eq</sub> for all granite samples were found to range from 100.5 to 517.6 Bq kg<sup>-1</sup> and the overall average value is 248.2 Bq kg<sup>-1</sup>. The obtained average value was much less than the assigned safe limit of 370 Bq kg<sup>-1</sup> as recommended by the Organization for Economic Cooperation and Development [20]. Accordingly, samples such as IGS<sub>2</sub>, IGS<sub>4</sub> and InGS<sub>7</sub> may pose radiation hazard.

The average values of D<sub>γ</sub> were found to vary from 48.5 to 343.6 nGy h<sup>-1</sup> with a total average value 118.2 nGy h<sup>-1</sup>. The obtained average value of D<sub>γ</sub> is 2.15 times higher than the world average value of 55 nGy h<sup>-1</sup> [4].

The EDR average values for samples under considerations were found to vary from 0.24 to 1.2 mSv year<sup>-1</sup>, with an overall average value of 0.58 mSv year<sup>-1</sup>, which is 1.41 times higher than the corresponding worldwide indoor value of 0.41 mSv year<sup>-1</sup> [2].

The calculated average values of H<sub>ex</sub> for the granite samples investigated in this study were found to range from 0.27 to 1.4 with a total average value of 0.67 (Table 3). Generally speaking, values of H<sub>ex</sub> of most surveyed samples were less than the safe limit (H<sub>ex</sub> < 1), except for IGS<sub>2</sub>, IGS<sub>4</sub> and InGS<sub>7</sub> samples.

The measured average values of the level index, I<sub>γ</sub>, were found to be higher than the international values (I<sub>γ</sub> > 1) for most samples. A few samples are found to have I<sub>γ</sub> ≤ 3 and I<sub>γ</sub> > 3. Such materials should be avoided according to recommendation of international foundations, since these values correspond to dose rates higher than 1 mSv year<sup>-1</sup> [2–4].

#### 4.3 Radon Exhalation Rate and the Effective Radium Content

The values of E<sub>x</sub> and C<sub>R</sub> are calculated using Eqs. (7)–(8) and the obtained results of are summarized in Table 4.

As it can be noticed from Table 4, E<sub>x</sub> average values varied from 3.9 to 30.5 Bqm<sup>-2</sup> day<sup>-1</sup> with an average value of 15.0 Bqm<sup>-2</sup> day<sup>-1</sup> in the granite samples collected from different stone saws in Palestinian regions. However, 43 % of the investigated granite samples had radon exhalation rates <10.0 Bqm<sup>-2</sup> day<sup>-1</sup>.

**Table 3** The average values of  $Ra_{eq}$ ,  $D_\gamma$ , EDR,  $H_{ex}$ , and  $I_\gamma$ , of the imported granite samples used in Palestinian regions

Sample code	$Ra_{eq}$ (Bq kg <sup>-1</sup> )	$D_\gamma$ (nGy h <sup>-1</sup> )	EDR (mSv year <sup>-1</sup> )	$H_{ex}$	$I_\gamma$
IGS <sub>1</sub>	366.6	174.3	0.86	0.99	2.7
IGS <sub>2</sub>	517.6	243.6	1.20	1.40	3.7
IGS <sub>3</sub>	237.7	111.4	0.55	0.64	1.7
IGS <sub>4</sub>	437.5	206.9	1.00	1.18	3.2
IGS <sub>5</sub>	209.7	102.3	0.50	0.57	1.6
IGS <sub>6</sub>	161.8	79.6	0.39	0.44	1.2
InGS <sub>7</sub>	435.8	206.2	1.00	1.18	3.4
InGS <sub>8</sub>	221.2	108.1	0.53	0.60	1.7
SGS <sub>9</sub>	220.1	106.2	0.52	0.59	1.6
SGS <sub>10</sub>	104.6	51.2	0.25	0.28	0.8
SGS <sub>11</sub>	120.4	57.2	0.28	0.33	0.9
BGS <sub>12</sub>	247.2	116.8	0.57	0.67	1.8
BGS <sub>13</sub>	100.5	48.5	0.24	0.27	0.7
ZGS <sub>14</sub>	132.5	63.5	0.31	0.36	1.0
Total average	248.2	118.2	0.58	0.67	1.8

**Table 4** The average values of track density,  $C_R$ , and  $E_x$  of radon of the imported granite samples used in Palestinian regions

Sample code	Av. track density (Tm <sup>-2</sup> ) × 10 <sup>3</sup>	$C_R$ (Bq kg <sup>-1</sup> )	$E_x$ (Bqm <sup>-2</sup> day <sup>-1</sup> )
IGS <sub>1</sub>	6.30	5.45	26.0
IGS <sub>2</sub>	7.70	6.12	30.5
IGS <sub>3</sub>	4.45	4.77	22.7
IGS <sub>4</sub>	7.12	6.00	27.7
IGS <sub>5</sub>	2.50	1.98	9.9
IGS <sub>6</sub>	2.30	2.10	9.7
InGS <sub>7</sub>	3.32	2.57	12.8
InGS <sub>8</sub>	2.65	2.15	10.5
SGS <sub>9</sub>	2.96	2.41	11.9
SGS <sub>10</sub>	0.98	0.81	3.9
SGS <sub>11</sub>	2.20	1.92	8.6
BGS <sub>12</sub>	5.41	4.30	21.5
BGS <sub>13</sub>	1.44	1.15	5.7
ZGS <sub>14</sub>	1.80	1.56	7.5
Total average	–	3.10	15.0

The average  $C_R$  values were found to vary from 0.81 to 6.12 Bq kg<sup>-1</sup> with a total average value of 3.1 Bq kg<sup>-1</sup>. The large variation in  $C_R$  values may be attributed to the variation of radium concentrations in each granite samples which are expected to have different effective radium content. Comparing the international value of 30 Bq kg<sup>-1</sup> [4] with the obtained results in the present work, it is clear that, except for one sample, radon concentrations are much smaller than the international average value.

**5 Conclusions**

Most selected granite samples had <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K average-specific activities greater than the world average values for building materials. The average activity concentrations

for most radionuclides varied with the production site of the granite, density and various colors. The average values of the activity concentrations in red and pink granites were higher than those in black and gray granites.

The average values of  $Ra_{eq}$ ,  $D_\gamma$ , EDR,  $H_{ex}$ ,  $I_\gamma$ ,  $E_x$ ,  $C_R$ , are found to be lower than the assigned international values except in four measurements. Such samples should be prevented from being used in construction purposes.

In conclusion, granite can be considered to be one of the main sources of radionuclides and radon exhalation in the environment. Based on the obtained data, some materials should be exempted from being used as building materials and decoration in Palestine. The obtained data may pave the way toward a baseline database of activity levels that can serve as a reference point for further radionuclide activity studies in Palestine.

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