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Radiological Hazard Assessment of Raw Granites from Ranyah, KSA

Sadek Zeghib*, Abdulkadir Sh. Aydarous, Ali Al-Qahtany

Environmental Protection Laboratory, Physics Department, Faculty of Science, Taif University, Taif, KSA
Email: *sadhmzag@hotmail.com

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Abstract

The assessment of radiological hazard due to external and internal indoor exposure was investigated for 26 raw granites collected from different granite quarries in Ranyah (KSA). The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured by high-resolution gamma spectrometry. Four granites were classified as "anomalous" due to their relatively high radioactivity. The averages and ranges of their activity concentrations were 667 (305 - 1120), 320 (161 - 491) and 586 (282 - 893) Bq·kg^{-1}, respectively. The corresponding ones for all remaining 22 granites were 45 (18 - 77), 39 (16 - 73) and 1178 (954 - 1531) Bq·kg^{-1}, respectively. In accordance with new European Basic Safety Standards (BSS) directives requiring a uniform reference level for indoor external exposure to gamma rays of 1 mSv·y^{-1}, all 22 granites may be used as bulk or ornamental building materials without any restrictions. Three anomalous granites should be subjected to control to be used as bulk materials. One anomalous granite was categorized as hazardous having an activity concentration index higher than 6. All four anomalous granites exceeded the level of newly adopted reference level of 300 Bq·m^{-3} for radon indoor exposure in case of poor ventilation. Two of them exceeded even for adequate ventilation.

Keywords

Environmental Radioactivity, Gamma Spectrometry, Activity Concentration, Radon, Granite

1. Introduction

Building materials can be the source of significant radiation exposure levels and give the most significant indoor gamma dose [1]. Granites, in particular, exhibit an enhanced elemental concentration of natural radionuclides in comparison to the very low abundance of these elements observed in the mantle and the crust of the Earth. The igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5 ppm of U and 15 ppm of Th), compared to rocks of basaltic or ultramafic composition (<1 ppm of U) [2] [3]. Granites mainly consist of coarse grains of quartz, potassium feldspar and sodium feldspar along with micas and hornblende as common minerals. Typical granites are chemically composed of 75% silica, oxides of aluminum, potassium and sodium at 12%, <5% and <5%, respectively, as well as smaller quantities of lime, iron, magnesia and titania [4].

Enhanced or elevated levels of Naturally Occurring Radioactive Material (NORM) in building materials for the construction of dwelling may cause effective doses, which exceeds the dose criterion of 1 $mSv \cdot y^{-1}$ [1] should be taken into account in terms of radiation protection. Terrestrial radiation contributes to external exposures from gamma radiation (outdoors and indoors), and to internal exposures from radon or dust radionuclides inhalation and ingestion. As an increasing concern about radiation risks from building materials, several principles, guidance and specific recommendations dealing with NORM were adopted [1] [5] [6]. Recently, the European Commission decided to harmonize, promote and consolidate these principles and recommendations, introducing them into the new EU directive laying down basic safety standards (new EU-BSS) for the protection against the danger arising from exposure to ionising radiation [6]. This directive was published in January 2014 adopting a uniform reference level of 1 $mSv \cdot y^{-1}$ for indoor external exposure to gamma rays emitted by building materials to identify those of concern from a radiation protection point of view. Before such materials are placed on the market, Member States are required to provide the radionuclides concentrations and the corresponding activity concentration index (ACI) as well as other relevant factors [6].

In Annex VIII of [6], it is stated that the activity concentration index value of 1 can be used as a conservative screening tool for identifying materials that may cause the reference level to be exceeded. In addition, the calculation of dose needs to take into account other factors such as density, thickness of the material as well as factors relating to the type of building and the intended use of the material (bulk or superficial). Many efforts led by different researchers have been focused on developing computational methodologies-room models- and *in situ* techniques to evaluate and predict the indoor gamma dose rate on the basis of the radioactivity and other characteristics of building materials [7]-[10].

The objective of this work is to assess the potential radiological risk to human health from 26 raw granites from Ranyah to be used eventually as building materials, in accordance with the new EU directive [6]. Similar studies were performed around the world. The gamma radiation in samples of a variety of natural tiling rocks (granites) imported in Cyprus for use in the building industry was measured, employing high-resolution γ-ray spectroscopy. The ranges of activity concentrations were determined for ^{232}Th (1 - 906 $Bq \cdot kg^{-1}$), ^{238}U (1 - 588 $Bq \cdot kg^{-1}$) and ^{40}K (50 - 1606 $Bq \cdot kg^{-1}$). Applying dose criteria recommended by the EU for superficial materials, 25 of the samples meet the exemption dose limit of 0.3 $mSv \cdot y^{-1}$, two of them meet the upper dose limit of 1 mSv·y^{-1} and only one clearly exceeds this limit [4].

A study was performed on some samples of marble and granite collected from different factories in Riyadh region of Saudi Arabia [11]. The measured values of the activities of ^{40}K, ^{226}Ra and ^{232}Th in the granite samples have been found to lie in the ranges: 0.28 - 1531.7, 0.03 - 147.0 and 0.02 - 186.4 Bq/kg, respectively. These samples were also found to have a radium equivalent activity in the range 0.089 - 504.61 Bq/kg. All the samples under investigation were found to have average external and internal hazard indices less than unity except the Brazilian granite sample.Similarly, twenty-four commercial granites sold in Saudi market (local and imported) were analyzed by [12]. The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in the selected granite samples ranged from 4.9 to 144, 9.7 to 133 and 168 to 1806 Bq·kg^{-1}, respectively. The radium equivalent activities (Ra_{eq}) are lower than the internationally accepted value limit of 370 Bq·kg^{-1} set by the Organization for Economic Cooperation and Development (OECD) [13], except in three imported granites.

In present work, the external gamma-ray dose rate was assessed in indoor environments covered with granites of 3 cm thickness for a standard room model considered by Anjos *et al*. [14] using Markkanen model code [7]. The latter consideredparameters like room size and building product through its density, thickness and composition to assess the indoor gamma dose rate due to building materials. Furthermore, the internal exposure was assessed through radon concentration using the same standard room model [14].

2. Materials and Methods

2.1. Samples Collection and Preparation for Laboratory Analysis

Ranyah province has attracted many companies for the exploitation of granites and ornamental stones, used as building materials that are sold locally and internationally. All the igneous rocks used as ornamental stone in the Kingdom are from the Proterozoic Arabian Shield. **Figure 1** shows the geological map of the area surrounding the Arabian shield along with Ranyah location [15]. A report was prepared by United States Geological Survey (USGS) about Precambrian geology of the Ranyah quadrangle [16]. They indicated that nearly all of the western half and the northeastern quarter of the quadrangle is underlied by intrusive granitic rock containing local residual blocks of older metabasalt and minor intrusive gabbro and felsic dikes. Perthite granite forms a prominent mountain range across the central part of the Ranyah quadrangle. Most of the granite is coarse grained and perthitic feldspar is ubiquitous. Some granites, especially near the edges of the mountain range and near the contact with the felsic volcanic rock are fine to medium grained. Approximately the northern half of the pluton is alkali granite [16] (and references included therein).

In this study, 26 raw granite samples were collected from different sectors of Ranyah near granite exploitation sites (quarries). The covered area was 35 km long and 15 km wide. A portable Scintillation Gamma Radiameter (SGR) was used to record the radiation

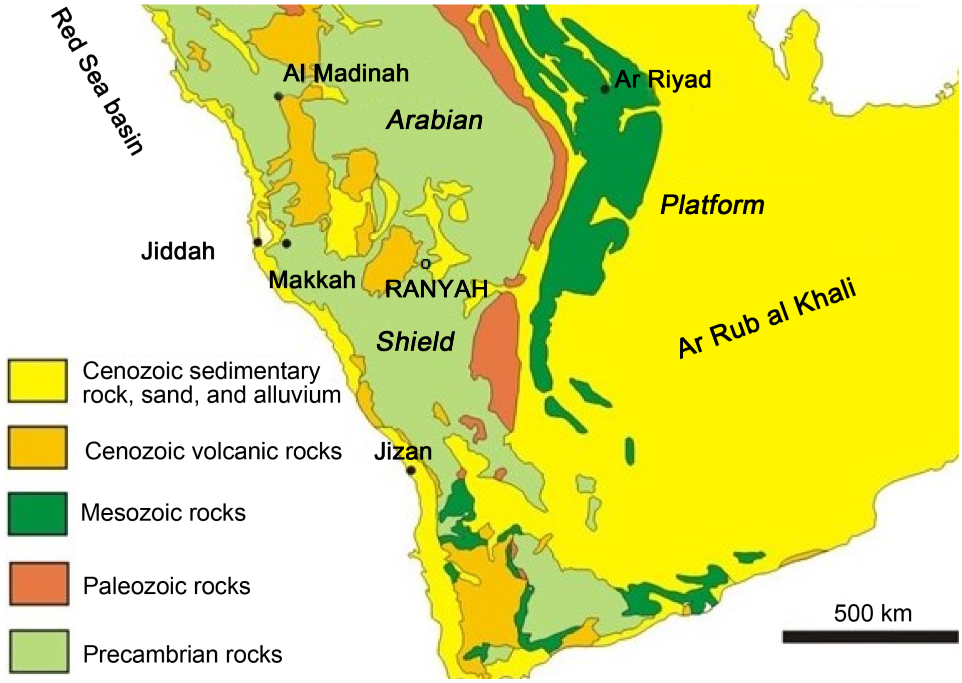

Figure 1. Geological map showing the main sedimentary rock main types in the eastern and the northern parts of the Kingdom of Saudi Arabia [15].

background level at different locations and to look for possible elevated radioactivity. Figure 2 shows the prominent mountain range across centrlal part of Ranyah along with several sampling locations. The four anomalous granites (AG1, AG2, AG3 and AG4) were collected from a different area called "Taghdoua". The granitic rock samples, each about 1.5 kg in weight, were first crushed, homogenized then sieved in order to have the same matrix as the reference sample. All samples were tightly sealed for a minimum of four weeks before measurements in 0.5 l Marinelli beakers, to reach secular equilibrium between ^{232}Th, ^{226}Ra and their short-lived progenies.

2.2. Gamma Rays Spectrometry Measurements

Measurement of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K was performed with high-resolution gamma ray spectrometry.A Canberra n-type hyper-pure Germanium (HPGe) detector (GR5021) of 50% relative efficiency was used for the analysis with a resolution of 2.1 keV and 1 keV (FWHM) for the 1.332 MeV gamma ray of ^{60}Co and 122 keV of ^{57}Co, respectively, equipped with model 747 Canberra lead shield system. The Data Acquisition system consists of Digital Spectrum Analyzer (DSA-2000), which is a fully integrated high performance multichannel analyzer. Analysis of spectra was performed by Genie 2000 software. The Standard radioactive source for efficiency calibration was prepared in the spectrometry laboratory at King Abdulaziz University using multi-gamma emitter ^{152}Eu. Such standard source was prepared in 0.5-liter Marinelli beakers having the same geometry as the samples. The 1460.8 keV line of Potassium ^{40}K was similarly considered in this respect. A full description of the procedure is

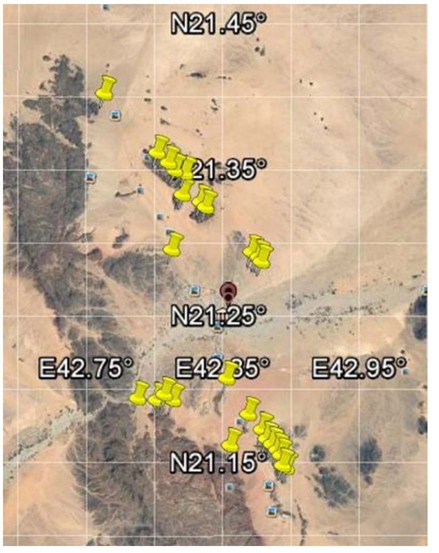

Figure 2. Satellite picture showing prominent mountain range across central part of Ranyah along with several sampling locations approximately.

given in [12]. The background spectrum was acquired for 864,000 s (24 hours) while all the samples were measured for 36,000 s. The ^{226}Ra activities were estimated from the gamma rays of ^{222}Rn decay products ^{214}Pb (295.2, 351.9 keV) and ^{214}Bi (609.3, 1120.3 keV). The ^{232}Th activities were estimated from the gamma rays of ^{212}Pb (238.6 keV), ^{228}Ac (338.4, 911.2 keV) and ^{208}Tl (583.2 keV). The ^{40}K activity was determined from its own gamma ray (1460.8 keV).

2.3. Assessment of Radiological Hazard

2.3.1. External Exposure to Gamma Radiation

External exposure is from gamma radiation emitted from ^{40}K, ^{226}Ra and ^{232}Th and their progenies and affects the whole body. The radiological hazard of granitic rocks used as building materials can be evaluated using the activity concentration index (ACI), proposed by the European Commission [17]:

$$\mathrm{ACI} = A_{\mathrm{Ra}}/300\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right) + A_{\mathrm{Th}}/200\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right) + A_{\mathrm{K}}/3000\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right) \quad (1)$$

where A_{Ra}, A_{Th} and A_{K} are the activity concentrations in Bq·kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The coefficients of ACI were calculated using a dose criterion of 1 mSv·year^{-1} exceeding the gamma dose received outdoors, *i.e.* 50 nGy·h^{-1}. As pointed out by Nuccetelli *et al.* [18], the activity concentration index should be used as a screening tool for identifying materials that may be exempted or subject to restrictions. For this purpose the activity concentration index I may be used for the classification of the materials into four classes, leading to two categories of building materials (A and B) according to **Table 1** following the radiological hazard classification defined in [17]. The division of materials into two other categories ((1) or (2)) according to their use shall be based on national building codes. Recommendations of EURATOM 2013 for building material require a uniform reference level for indoor external exposure to

Table 1. Categories based on the default dose according to the ACI criteria defined in [17].

Use of materials	Category (corresponding default dose)	
	A (≤1 mSv)	B (>1 mSv)
1) in bulk amounts	A1 (I ≤ 1)	B1 (I > 1)
2) Superficial and/or with restricted use	A2 (I ≤ 6)	B2 (I > 6)

gamma rays of 1 mSv·y^{-1} [6]. Adopting the conversion factor from the absorbed dose in air to effective dose received by adults (0.7 Sv·Gy^{-1}) and the indoor occupancy factor (0.8) proposed by UNSCEAR (2000) [1], the annual effective dose rate indoors (E_{ind}) is calculated using the following formula:

$$E_{ind}\left(\text{mSv}\cdot\text{y}^{-1}\right) = D_{ind}\left(\text{nGy}\cdot\text{h}^{-1}\right)\times 8760\left(\text{h}\cdot\text{y}^{-1}\right)\times 0.8\times 0.7\left(\text{Sv}\cdot\text{Gy}^{-1}\right)\times 10^{-6} \quad (2)$$

where D_{ind} (nGy·h^{-1}) is the absorbed gamma dose rate in indoor air due to external exposure from gamma radiation from building materials of dwelling. Different models have been adopted for a standard room and its configuration, to evaluate it. The indoor exposure to external gamma radiation depends on the form of the dwelling, the properties of building materials (density, thickness and elemental composition) and of course on the activity concentrations of NORM in these building materials. Researchers [7] [8] have adopted Monte Carlo simulations to obtain the free-in-air dose rate resulting from gamma rays emitted from the floor, walls and ceiling of a standard room with specific dimensions. It has been reported that specific dose rates depended to a large degree on wall thickness and density but not so on position in the room and dimensions of the room [8]. The same model room 1 for the configuration of standard rooms (4.0 m × 5.0 m area and 2.8 m high), in which the walls and floor are covered with granite slabs of 3.0 cm thickness as adopted by Anjos *et al.* [14], was considered. As noted, the specific dose rates were calculated with a computer program published by Markkanen assuming a density of 2600 kg·m^{-3} [7]. It is worth noting that similar configurations are extensively used in KSA (dwellings, hospitals, clinics, restaurants …etc.). The free-in-air gamma dose rate caused by walls and floor in the middle of the room was given in terms of the values of the activity concentrations A_{Ra}, A_{Th} and A_K by [14]:

$$D_{ind}\left(\text{nGy}\cdot\text{h}^{-1}\right) = 0.17A_{Ra} + 0.20A_{Th} + 0.013A_K \quad (3)$$

2.3.2. Internal Exposure to Radon Gas

Internal exposure due to the intake of radionuclide through inhalation of indoor radon gas is harmful to health when it exceeds the limits. Radon progenies are solid radioactive elements that are deposited in the respiratory tract tissues, which may lead to lung cancer. The radon concentration in dwellings depends on many factors such as the room model, the ventilation, the nature, type, and amount of building materials as well as the way they are used. Therefore, a more realistic assessment of the internal exposure to radon gas (^{222}Rn) is preferred, instead of just relying on the usual estimation of the internal hazard index H_{in} or on the alpha index I_α. Recent regulations lead to the establishment of a new national reference level ≤300 Bq·m^{-3} for radon in dwellings and workplaces [6]. It is recommended that existing dwellings exceeding the reference level should be identified and encouragement of radon-reducing measures be implemented where necessary.

In view of the relatively high ^{226}Ra activity concentrations of anomalous granites (AG), the exposure to radon by estimating the concentration was assessed more realisticallyin the same standard room model assumed before. It is given by the following formula [14]:

$$C_{\text{Rn}} = \frac{\frac{E_x S}{V} + C_0 \lambda_v}{(\lambda + \lambda_v)} \tag{4}$$

where E_x is the exhalation rate per unit area, C_o is the radon concentration (Bq·m^{-3}) of the outside air, λ_v is the air removal rate due to ventilation (h^{-1}), and λ is the decay constant of radon (7.54 × 10^{-3} h^{-1}). S is the exhaling surface area (m^2) and V is the volume of the room (m^3). The exhalation rate per unit area, originating from the walls and floor covered with different types of granite was calculated theoretically according to the following formula (for dry condition) by [19]:

$$E_x = \frac{1}{2} A_{\text{Ra}} \lambda \rho \eta d \tag{5}$$

where ρ is the material density (assumed to be 2600 kg·m^{-3}), d is the wall thickness (m), and η is the emanation coefficient, *i.e.* the fraction of radon that reaches to the wall surface by diffusion process. The same parameters (C_o = 10 Bq·m^{-3}, d = 3 cm^3, η = 0.45) considered by Anjos *et al.* [14] (and references included therein) for model room 1, were used in our calculation too. Likewise, the ratio of the exhaling area covered with granite slabs to the free room volume was assumed to be S/V = 2.0 m^{-1} (considering that part of the room volume is occupied by furniture). In addition, for a safe assessment, the maximum measured value η = 0.45 for radon emanation coefficient for granites used in Saudi Arabia [20] was adopted in the calculation [14].

According to UNSCEAR [19] reports, λ_v values varies between 0.1 h^{-1} and 3 h^{-1} for residence. Value of λ_v < 0.1 h^{-1} are for extremely poor ventilation cases. An air exchange rate of λ_v = 0.5 h^{-1} was suggested for residential mechanical systems, which was considered for an adequately ventilated room [14]. The calculation was performed for these two values for all granite samples.

3. Results and Discussion

3.1. External Exposure

The calculated indoor absorbed gamma dose rate, D_{ind} and annual effective dose rate, E_{ind} for anomalous granites due to external gamma exposure when used as superficial building material in the previously specified room model are given in **Table 2**. The arithmetic mean and the standard deviation (σ) are given for normal (NG) and anomalous (AG) granites separately. The last column indicates the classification of each

Table 2. Activity concentrations and radiological quantities for normal and anomalous raw granites.

Sample	^{232}Th (Bq·kg^{-1})	^{226}Ra (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})	Ra_{eq} (Bq·kg^{-1})	D_{ind} (nGy·h^{-1})	E_{ind} (mSv·y^{-1})	ACI	Category
NG1	39.8 ± 2.8	59.3 ± 3.2	1245 ± 6	212.1	34.2	0.17	0.81 ± 0.02	A1, A2
NG2	37.2 ± 1.9	33.2 ± 1.7	1295 ± 5	186.2	29.9	0.15	0.73 ± 0.01	A1, A2
NG3	34.0 ± 1.0	39.7 ± 2.8	1243 ± 5	184	29.7	0.15	0.72 ± 0.01	A1, A2
NG4	15.7 ± 0.7	22.3 ± 1.3	1220 ± 5	138.6	22.8	0.11	0.56 ± 0.01	A1, A2
NG5	33.8 ± 1.8	42.9 ± 2.7	1220 ± 5	185.2	29.9	0.15	0.72 ± 0.01	A1, A2
NG6	17.0 ± 1.1	18.1 ± 0.7	1531 ± 6	160.2	26.4	0.13	0.66 ± 0.01	A1, A2
NG7	41.2 ± 2.3	42.2 ± 2.6	1005 ± 5	178.4	28.5	0.14	0.68 ± 0.01	A1, A2
NG8	62.8 ± 2.7	64.2 ± 4.2	1065 ± 5	236	37.3	0.18	0.88 ± 0.02	A1, A2
NG9	35.8 ± 0.8	46.7 ± 1.9	1249± 6	194.1	31.3	0.15	0.75 ± 0.01	A1, A2
NG10	39.2 ± 1.6	49.0 ± 2.5	1251 ± 5	201.3	32.4	0.16	0.78 ± 0.01	A1, A2
NG11	36.2 ± 2.0	30.3 ± 1.2	1135 ± 5	169.4	27.1	0.13	0.66 ± 0.01	A1, A2
NG12	34.1 ± 2.4	24.0 ± 1.3	1298 ± 6	172.8	27.8	0.14	0.68 ± 0.01	A1, A2
NG13	39.9 ± 1.6	36.7 ± 1.7	1318 ± 5	195.1	31.3	0.15	0.76 ± 0.01	A1, A2
NG14	45.5 ± 2.6	49.7 ± 3.0	1097 ± 5	199.2	31.8	0.16	0.76 ± 0.02	A1, A2
NG15	50.6 ± 2.8	50.2 ± 2.7	954 ± 5	196.1	31.1	0.15	0.74 ± 0.02	A1, A2
NG16	72.5 ± 3.8	71.1 ± 4.6	1112 ± 5	260.4	41.0	0.20	0.97 ± 0.02	A1/B1, A2
NG17	47.5 ± 1.5	49.2 ± 2.6	1060 ± 5	198.8	31.6	0.16	0.75 ± 0.01	A1, A2
NG18	41.0 ± 1.6	77.4 ± 3.4	1132 ± 5	223.2	36.1	0.18	0.84 ± 0.01	A1, A2
NG19	43.7 ± 2.0	49.0 ± 2.9	1154 ± 5	200.3	32.1	0.16	0.77 ± 0.01	A1, A2
NG20	21.6 ± 1.2	21.7 ± 1.9	1177 ± 5	143.2	23.3	0.11	0.57 ± 0.01	A1, A2
NG21	22.8 ± 1.1	31.4 ± 1.9	1064 ± 5	145.9	23.7	0.12	0.57 ± 0.01	A1, A2
NG22	44.1 ± 1.1	74.5 ± 4.0	1093 ± 5	221.7	35.7	0.18	0.83 ± 0.01	A1, A2
Mean ± σNG	39 ± 13	45 ± 17	1178 ± 127	191.0 ± 30	30.7 ± 4.6	0.15 ± 0.02	0.74 ± 0.10	
AG1	394 ± 11	833 ± 44	681 ± 5	1448.3	229.2	1.13	4.97 ± 0.16	A2, B1
AG2	491± 17	1120 ± 59	486 ± 6	1858.9	294.8	1.45	6.35 ± 0.21	B2
AG3	232 ± 8	410 ± 23	282 ± 4	762.4	119.6	0.59	2.62 ± 0.09	A2, B1
AG4	161 ± 7	305 ± 17	893 ± 5	603.6	95.6	0.47	2.12 ± 0.07	A2, B1
Mean ± σAG	320 ± 150	667 ± 379	586 ± 262	1168.3 ± 588	185 ± 94	0.91 ± 0.46	4.02 ±1.99	

granite according to **Table 1**. It is clear that samples AG1 and AG2 exceeded the reference level for indoor external exposure to gamma rays of 1 mSv·y^{-1} with 1.13 and 1.45 mSv·y^{-1} values, respectively. The mean value and the standard deviation for all anomalous granites (0.91 ± 0.46 mSv·y^{-1}) are much bigger than those for normal granites (0.15 ± 0.02 mSv·y^{-1}). Based on the activity concentration indices, all NG samples except one classified as "normal" were categorized as A1 and A2 having ACI < 1 (suitable for being used as bulk and surface materials without restriction). Normal granite NG16 was classified as A1/B1to indicate a potential radiological hazard in bulk utilization of this granite, and should be subject to control (having ACI-1 within the computed uncertainty).

Three out of four anomalous granites were categorized as B1 having ACI > 1 (restricting their use as bulk materials and should be subject to control). However, they can be used superficially without restrictions having ACI < 6 (A2 category). Anomalous granite AG2 was categorized as B2 material forbidding completely its use as construction material having ACI > 6 (I = 6.35). In column 5 of **Table 2**, the radium equivalent activity (Ra_{eq}) defined by Beretka and Mathew [21], has been included to show that all four anomalous granites far exceed the permissible value from building materials of 370 Bq·kg^{-1} with an average value of 1168 Bq·kg^{-1} and a range extending from 604 to 1859 Bq·kg^{-1}.

It is remarkable that most of the granite samples with high radioactivity examined by Chen and Lin [22] were also red coloured as samples AG1 and AG2. The red colour of their highly radioactive granites (Balmoral and African red) is due to the presence of abundant feldspars that are reddish in colour as reported by Pavlidou *et al.* [23]. However, all four "anomalous" granites were collected from a different region where the small rocky mountain of the anomaly area is not subject to exploitation.

Trevisi *et al.* [24] noted in their database, that among the 621 superficial materials used in 15 MS (Member States) of EU (European Union), only two had ACI (I_{max}) higher than 6 (Italian basalt I ≈ 6.10 and a granite, commercially named as *Café Brown,* imported to Greece I ≈ 7.03). In our present study, one out of twenty six (granite AG2) had exceeded this critical value for superficial building materials. **Table 3** shows a comparison between the results from the EU database, few recent studies worldwide

Table 3. Averages and ranges for activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K for stones used as superficial material in EU member states and a comparison with present study and few recent ones worldwide.

Superficial stones	No. of samples	^{226}Ra (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})	Country [Ref.]
Igneous plutonic	387	78 (0.8 - 588)	89 (0.3 - 906)	1049 (24 - 2040)	European Union [17]
Igneous volcanic	86	160 (16 - 709)	163 (8 - 750)	1295 (170 - 2354)	
Metamorphic	148	27 (0.7 - 166)	21 (0.0 - 142)	395 (0.2 - 1891)	
Granites[a]		(4.9 - 190)	(4.5 - 450)	(190 - 2029)	Brazil [14]
Volcanic tuff stones[a]	76	(2 - 263)	(8 - 401)	(99 - 2107)	Turkey [25]
Granites	50	47 (17 - 85)	83 (62 - 114)	1426 (1315 - 1551)	Nigeria [26]
Granitoid outcrops[b]	7	44 (29 - 53)	56 (51 - 60)	1133 (711 - 1355)	Italy [9]
Granites		67 (2 - 95)	95 (1 - 450)	1200 (50 - 3800)	Greece [27]
Granites		187 (80 - 330)	118 (100 - 140)	852 (250 - 1300)	Egypt [28]
Granites	20	659 (46 - 6180)	598 (92 - 3214)	1218 (899 - 1987)	Pakistan [29]
Ranyah Anomalous raw Granites	4	667 (305 - 1120)	319 (161 - 491)	586 (282 - 893)	Saudi Arabia [Present Work]
Ranyah Normal raw Granites	22	45 (18 - 77)	39 (16 - 73)	1178 (954 - 1531)	

[a]Average values not available. [b]HPGe measurements were considered.

and the measurements for our normal and anomalous raw granites, which were collected from Ranyah region.

Other "anomalies" with much higher level of activity concentration values were found in young granites in the Egyptian desert at "Um Taghir". The average values of ^{226}Ra, ^{232}Th and ^{40}K activity concentration were 3732, 1683 and 4801 $Bq \cdot kg^{-1}$, respectively [30].

3.2. Internal Exposure to Radon Gas

As mentioned early, the calculations for radon concentration were performed for two values for the air removal rate due to ventilation: λ_v = 0.1 h^{-1} for poor ventilation and λ_v = 0.5 h^{-1} for adequate ventilation. The results are shown in **Figure 3** and **Figure 4** for all samples including the mean value for normal granites (MNG). It is worth mentioning that the reference level of 300 $Bq \cdot m^{-3}$ for radon in dwellings represents approximately 10 $mSv \cdot y^{-1}$ [31]. It is clear that all 22 granites classified as "normal", are safe in both circumstances. For a poor ventilated condition, all four anomalous granites clearly exceed the reference level. However, when the ventilation is adequate, only samples AG1 and AG2 exceed this limit and should not be used as superficial building materials (same red colour in both figures). By comparison, Anjos *et al.* [14] found that all their investigated granites had a value below 100 $Bq \cdot m^{-3}$ in case of adequate ventilation. In case of poor ventilation, 9% of 71 analysed Brazilian commercial granites showed a value above 300 $Bq \cdot m^{-3}$ with a maximum value of (~400 $Bq \cdot m^{-3}$). In our case, the maximum value (~2800 $Bq \cdot m^{-3}$) is seven times higher in case of poor ventilation due to the relatively high ^{226}Ra activity concentrations in anomalous granites.

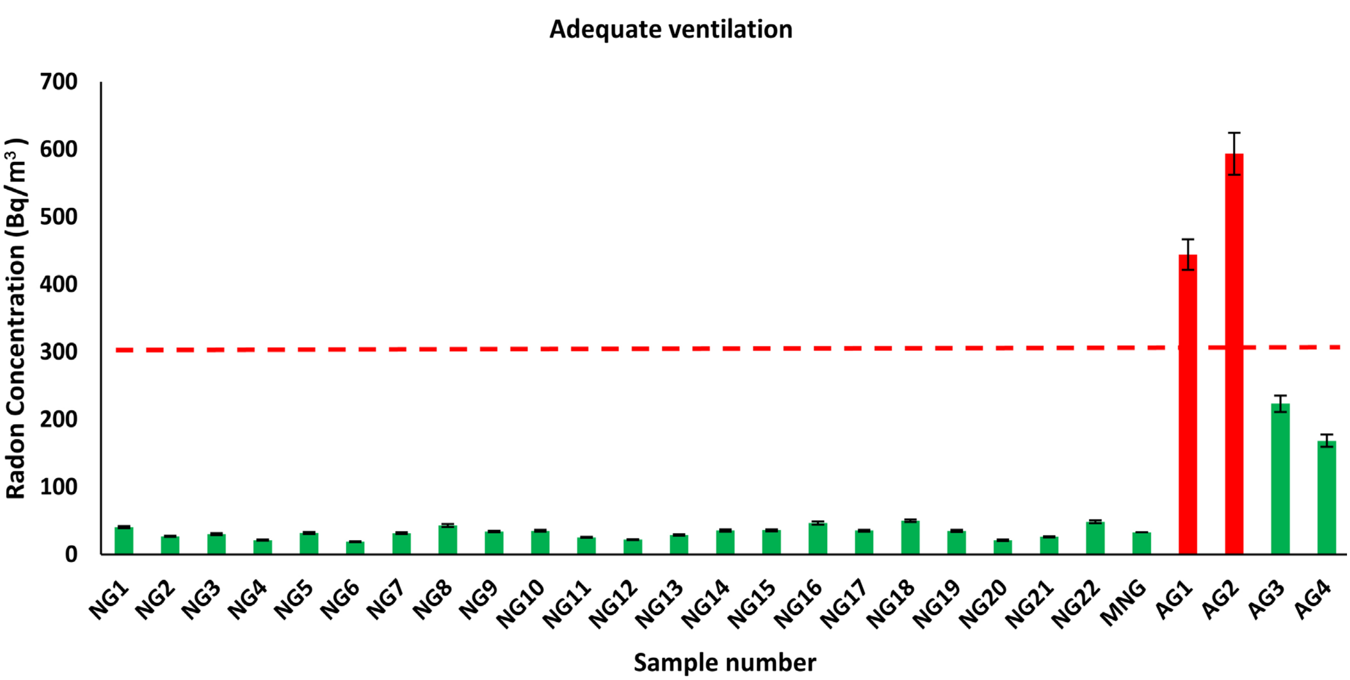

Figure 3. Radon concentration for adequately ventilated room (λ_v = 0.5 h^{-1}) (Reference level in broken line).

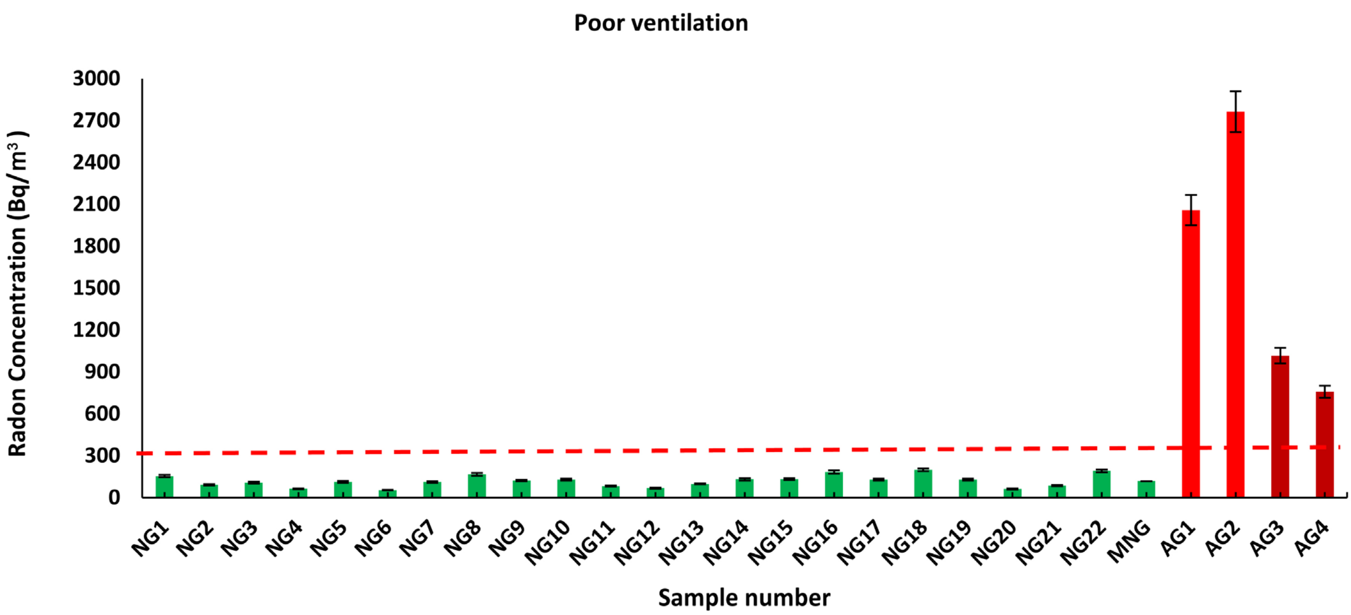

Figure 4. Radon concentration for poorly ventilated room ($\lambda_v = 0.1\ h^{-1}$) (Reference level in broken line).

3.3. Multi-Elemental Analysis

The collected anomalous granites are characterized by their high ^{226}Ra and ^{232}Th activity concentrations. At the same time, they have relatively low ^{40}K activity concentration by comparison with other collected granites. A recent work with plutonic bodies used as decorative building material in Greece investigated to correlate the mineralogical composition, the chemical composition (major oxides), groups of minerals and ratios of major oxides, age and grain size with ^{226}Ra and ^{232}Th activities [32]. However, obvious correlations were not found as the R^2 values were below 0.1 in all cases, even for SiO_2. Therefore, we restricted our analysis to the four anomalous granites along with granite NG6 (which has the highest ^{40}K activity concentration (1531 Bq·kg^{-1}) to look for possible correlation with their elemental composition. Energy dispersive X-ray Spectrometry (EDS) was conducted at Taif University JEOL scanning electron microscope (JEOL SEM 6390 LA). An example of SEM image and associated standard-less EDS spectrum is shown in Figure 5 for hazardous graniteAG2 that had the highest level of natural radioactivity in our present work. Very similar spectra were obtained for other samples. Silicon (Si) is the dominant element in all samples. Other common elements are potassium (K), aluminium (Al), Sodium (Na), Calcium (Ca), iron (Fe) and Magnesium (Mg). In addition, Titanium Ti is found in granite NG6 in small amount whereas as traces in others. These elements belong to the following oxides found in general in granites as pointed out by Krmar *et al.* [33]: SiO_2, K_2O, Al_2O_3, Na_2O, CaO, Fe_2O_3 and MgO. The final quantitative analysis of all four anomalous granites along with granite NG6 are summarized in Table 4, where the mass percentages of different elements present in the samples are shown. The correlation between K content and ^{40}K activity concentration is obvious. The correlation coefficient R^2 reached a high value of 0.95 as shown in Figure 6.

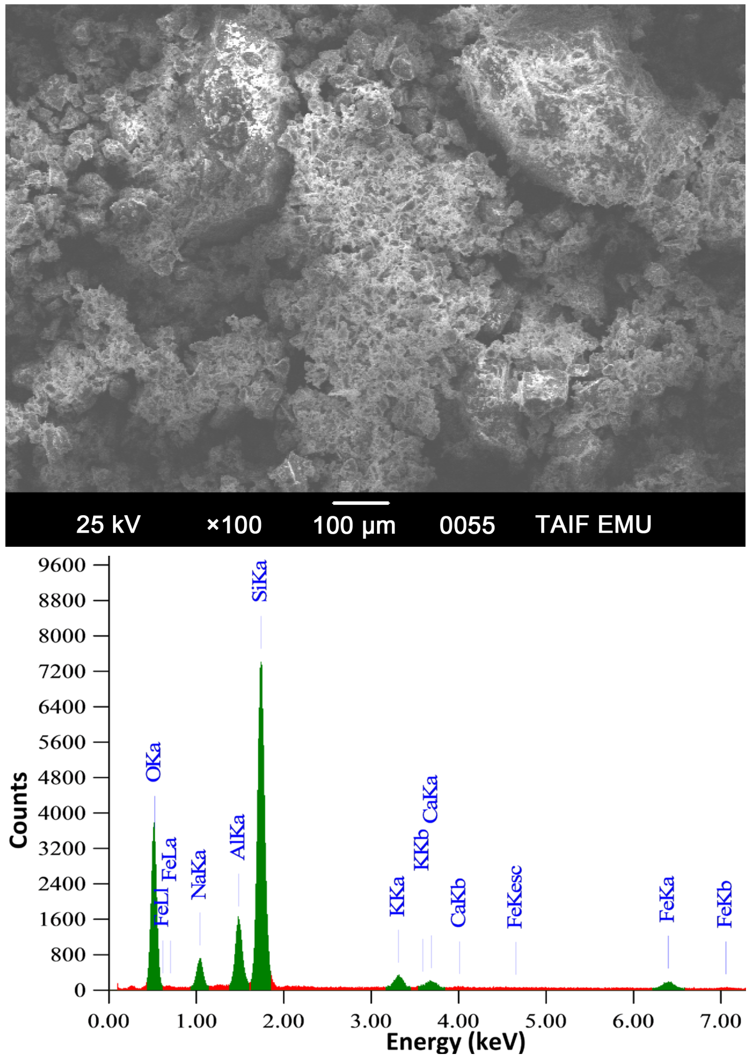

Figure 5. EDS spectrum (down) and corresponding SEM image (up) for most radioactive anomalous granite AG2.

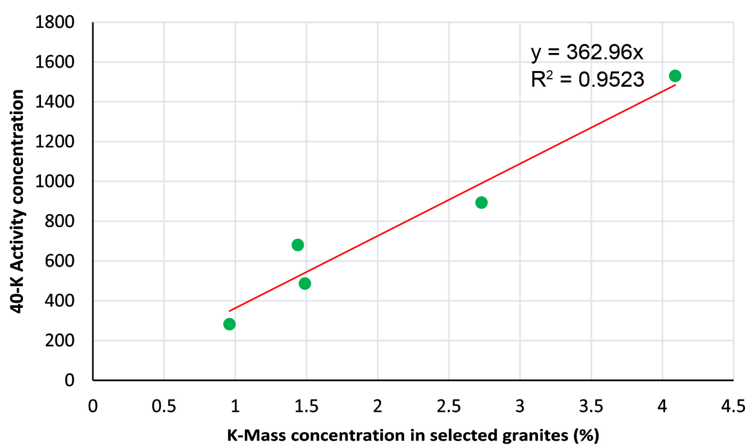

Figure 6. Correlation between K-Mass concentration and ^{40}K Activity concentration for AG1, AG2, AG3, AG4 and NG6 granites.

Table 4. EDS multi-elemental Analysis-Mass percentage (%).

Element	AG1	AG2	AG3	AG4	NG6
O	53.12	52.46	52.33	52.63	50.90
Na	4.81	5.07	4.00	3.60	2.60
Mg	trace	trace	trace	trace	1.03
Al	6.49	6.84	5.39	6.52	7.81
Si	28.09	31.23	32.32	29.90	26.36
K	1.44	1.49	0.96	2.73	4.09
Ca	3.32	0.79	2.14	2.64	2.68
Fe	2.73	2.11	2.86	1.99	4.54

4. Conclusion

All twenty-two raw granites collected from areas near quarries are safe to be used as building materials without any restriction. Three out of four anomalous granites collected from "Taghdoua" have restricted use and should be subject to control. Anomalous granite AG2 was categorized as B2 material forbidding completely its use as construction material as far as external exposure to gamma dose is concerned. Although radon indoor exposure can be significantly reduced through adequate ventilation, anomalous granites AG1 and AG2 remain unsafe as superficial building materials for the standard room considered because of their relatively high ^{226}Ra activity concentrations. It is recommended that companies have to check the radiation risk of granites intended for commercialization before any large-scale exploitation using appropriate instruments. Besides, the importance of the present work for environmental radiological protection in general, it will serve as a valuable information for the current Saudi Geological Survey strategic programs in environmental geology that are concentrated on mapping the hazards associated with natural radioactivity.

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References

[1] UNSCEAR (2000) United Nations Scientific Committee on the Effects of Atomic Radiation, Sources, Effects and Risks of Ionizing Radiation, Report to General Assembly, with Scientific Annex B: Exposure from Natural Sources of Radiation, United Nations, New York.

[2] Faure, G. (1986) Principles of Isotope Geology. John Wiley & Sons, New York.

[3] Menager, M.T., Heath, M.J., Ivanovich, M., Montjotin, C., Barillon, C.R., Camp, J. and Hasler, S.E. (1993) Migration of Uranium from Uranium-Mineralised Fractures into the Rock Matrix in Granite: Implications for Radionuclide Transport around a Radioactive Waste Repository. 4*th International Conference of Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere* (*Migration* 1993), Charleston, 12-17 December 1993, Radiochimica Acta 66/67, 47-83.

[4] Tzortzis, M., Tsertos, H., Christofides, S. and Christodoulides, G. (2003) Gamma Radiation

Measurements and Dose Rates in Commercially Used Natural Tiling Rocks (Granites). *Journal of Environmental Radioactivity*, **70**, 223-235. http://dx.doi.org/10.1016/S0265-931X(03)00106-1

[5] European Commission (1999) Nuclear Safety and Civil Protection. Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials. Radiation Protection 112.

[6] Council of the European Union (2014) Council Directive 2013/59/EURATOM of 5 December 2013 Laying Down Basic Safety Standards for Protection against the Dangers Arising from Exposure to Ionising Radiation. Off. J. Eur. Union L 13, Brussels, 1e73.

[7] Markkanen, M. (1995) Radiation Dose Assessments for Materials with Elevated Natural Radioactivity, Report STUK-B-STO 32. Radiation and Nuclear Safety Authority-STUK.

[8] Risica, S., Bolzan, C. and Nuccetelli, C. (2001) Radioactivity in Building Materials: Room Model Analysis and Experimental Methods. *Science of the Total Environment*, **272**, 119-126. http://dx.doi.org/10.1016/S0048-9697(01)00675-1

[9] Puccini, A., Xhixha, G., Cuccuru, S., Oggiano, G., Xhixha, M.K., Mantovani, F., Alvarez, C.R. and Casini, L. (2014) Radiological Characterization of Granitoid Outcrops and Dimension Stones of the Variscan Corsica-Sardinia Batholith. *Environmental Earth Sciences*, **71**, 393-405. http://dx.doi.org/10.1007/s12665-013-2442-8

[10] Nuccetelli, C., Leonardi, F. and Trevisi, R. (2015) A New Accurate and Flexible Index to Assess the Contribution of Building Materials to Indoor Gamma Exposure. *Journal of Environmental Radioactivity*, **143**, 70-75. http://dx.doi.org/10.1016/j.jenvrad.2015.02.011

[11] Al-Saleh, F.S. and Al-Berzan, B. (2007) Measurements of Natural Radioactivity in Some Kinds of Marble and Granite Used in Riyadh Region. *Journal of Nuclear and Radiation Physics*, **2**, 25-36.

[12] Aydarous, A.S., Zeghib, S. and Al-Dughmah, M. (2010) Measurements of Natural Radioactivity and the Resulting Radiation Doses from Commercial Granites. *Radiation Protection Dosimetry*, **142**, 363-368. http://dx.doi.org/10.1093/rpd/ncq216

[13] OECD (1979) Organization for Economic Cooperation and Development. Exposure to Radiation from the Natural Radioactivity in Building Materials. Report by a Group of Experts of the OECD, Paris.

[14] Anjos, R.M., Juri Ayub, J., Cid, A.S., Cardoso, R. and Lacerda, T. (2011) External Gamma-Ray Dose Rate and Radon Concentration in Indoor Environments Covered with Brazilian Granites. *Journal of Environmental Radioactivity*, **102**, 1055-1061. http://dx.doi.org/10.1016/j.jenvrad.2011.06.001

[15] http://www.sgs.org.sa/English/Geology/Phanerozoic/Pages/default.aspx. (Accessed on August 10th, 2016)

[16] Fenton, D. (1983) The Mineral Resource Potential of the Harrat Nawasif, Sheet 21/42 C, Ranyah, Sheet 21/42 D and Jabal Dalfa, Sheet 21/43 C, Quadrangles, Kingdom of Saudi Arabia. Open-File Report, USGS-OF-1983-0372. Ministry of Petroleum and Mineral Resources Deputy, Jiddah, Kingdom of Saudi Arabia. https://pubs.usgs.gov/of/1983/0372/report.pdf

[17] European Commission (EC) (2011) Draft Presented under Article 31Euratom Treaty for the Opinion of the European Economic and Social Committee: Laying down Basic Safety Standards Forprotection against the Dangers Arising from Exposure to Ionizing Radiation. COM 593, Brussels,

[18] Nuccetelli, C., Risica, S., D'Alessandro, M. and Trevisi, R. (2012) Natural Radioactivity in Building Material in the European Union: Robustness of the Activity Concentration Index I and Comparison with a Room Model. *Journal of Radiological Protection*, **32**, 349-358.

http://dx.doi.org/10.1088/0952-4746/32/3/349

[19] UNSCEAR (1988) United Nations Scientific Committee on the Effects of Atomic Radiation, Sources and Effects of Ionizing Radiation. Report to General Assembly, with Annexes, United Nations, New York.

[20] Al-Jarallah, M. (2001) Radon Exhalation from Granites Used in Saudi Arabia. *Journal of Environmental Radioactivity*, **53**, 91-98. http://dx.doi.org/10.1016/S0265-931X(00)00110-7

[21] Beretka, J. and Mathew, P.J. (1985) Natural Radiactivity of Australian Building Materials, Industrial Wastes and By-Product. *Health Physics*, **48**, 87-95. http://dx.doi.org/10.1097/00004032-198501000-00007

[22] Chen, C.J. and Lin, Y.M. (1995) Assessment of Building Materials for Compliance with Regulations of ROC. *Environment International*, **22**, 221-226. http://dx.doi.org/10.1016/S0160-4120(96)00111-0

[23] Pavlidou, S., Koroneos, A., Papastefanou, C., Christofides, G., Stoulos, S. and Vavelides, M. (2006) Natural Radioactivity of Granites Used as Building Materials. *Journal of Environmental Radioactivity*, **89**, 48-60. http://dx.doi.org/10.1016/j.jenvrad.2006.03.005

[24] Trevisi, R., Risica, S., D'Alessandro, M., Paradiso, D. and Nuccetelli, C. (2012) Natural Radioactivity in Building Materials in the European Union: A Database and an Estimate of Radiological Significance. *Journal of Environmental Radioactivity*, **105**, 11-20. http://dx.doi.org/10.1016/j.jenvrad.2011.10.001

[25] Turhan, Ş., Atıcı, E. and Varinlioğlu, A. (2015) Radiometric Analysis of Volcanic Tuff Stones Used as Ornamental and Structural Building Materials in Turkey and Evaluation of Radiological Risk. *Radioprotection*, **50**, 273-280. http://dx.doi.org/10.1051/radiopro/2015020

[26] Stoulos, S., Manolopoulou, M. and Papastefanou, C. (2003) Assessment of Natural Radiation Exposure and Radon Exhalation from Building Materials in Greece. *Journal of Environmental Radioactivity*, **69**, 225-240. http://dx.doi.org/10.1016/S0265-931X(03)00081-X

[27] Ahmed, N.K. (2005) Measurement of Natural Radioactivity in Building Materials in Qena City, Upper Egypt. *Journal of Environmental Radioactivity*, **83**, 91-99. http://dx.doi.org/10.1016/j.jenvrad.2005.03.002

[28] Ademola, J.A. and Ayeni, A.A. (2010) Measurement of Natural Radionuclides and Dose Assessment of Granites from Ondo State, Nigeria. *Radioprotection*, **45**, 513-521. http://dx.doi.org/10.1051/radiopro/2010046

[29] Asghar, M., Tufail, M., Sabiha-Javied Abid, A. and Waqas, M. (2008) Radiological Implications of Granite of Northern Pakistan. *Journal of Radiological Protection*, **28**, 387-398. http://dx.doi.org/10.1088/0952-4746/28/3/009

[30] El-Arabi, A.M. (2007) ^{226}Ra, ^{232}Th and ^{40}K Concentrations in Igneous Rocks from Eastern Desert Egypt and Its Radiological Implications. *Radiation Measurements*, **42**, 94-100. http://dx.doi.org/10.1016/j.radmeas.2006.06.008

[31] ICRP (2009) International Commission on Radiological Protection. Statement on Radon, ICRP Ref 00/902/09.

[32] Papadopoulos, A., Christofides, G., Koroneos, A., Papadopoulou, L., Papastefanou, C. and Stoulos, S. (2013) Natural Radioactivity and Radiation Index of the Major Plutonic Bodies in Greece. *Journal of Environmental Radioactivity*, **124**, 227-238. http://dx.doi.org/10.1016/j.jenvrad.2013.06.002

[33] Krmar, M., Slivka, J., Varga, E., Bikit, I. and Veskovic, M. (2009) Correlation of Natural Radionuclides in Sediment from Danube. *Journal of Geochemical Exploration*, **100**, 20-24. http://dx.doi.org/10.1016/j.gexplo.2008.03.002