

PERIODICO di MINERALOGIA  
established in 1930

*An International Journal of  
MINERALOGY, CRYSTALLOGRAPHY, GEOCHEMISTRY,  
ORE DEPOSITS, PETROLOGY, VOLCANOLOGY*  
and applied topics on *Environment, Archaeometry and Cultural Heritage*

## Natural radioactivity and dose assessment of granitic rocks from the Atticocycladic Zone (Greece)

Argyrios Papadopoulos<sup>1,\*</sup>, Georgios Christofides<sup>1</sup>, Antonios Koroneos<sup>1</sup>,  
Stylianou Stoulos<sup>2</sup> and Constantinos Papastefanou<sup>2</sup>

<sup>1</sup>Department of Mineralogy, Petrology and Economic Geology, School of Geology  
Aristotle University of Thessaloniki, 541 24 Thessaloniki, Greece

<sup>2</sup>Laboratory of Atomic and Nuclear Physics, School of Physics,  
Aristotle University of Thessaloniki, 541 24 Thessaloniki, Greece

\*Corresponding author: [argpapad@geo.auth.gr](mailto:argpapad@geo.auth.gr)

### Abstract

The activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K of granite samples taken from the Atticocycladic Zone were measured by gamma-ray spectrometry. These concentrations were compared to the commercial granites imported in Greece and Cyprus. The absorbed dose rate, the annual effective dose and the gamma-ray index were determined, so as to assess the radiological impact from the granites investigated, in case they were used as building materials. The range of the absorbed dose rate, the annual effective dose and the gamma index was around a mean value of  $86 \pm 30$  nGy h<sup>-1</sup>,  $0.6 \pm 0.2$  mSv y<sup>-1</sup> and  $1.0 \pm 0.2$ , respectively. Taking into consideration the internal exposure due to radon inhalation, the annual effective dose, in a room fully constructed of granite, varies between 0.4 and 1.4 mSv y<sup>-1</sup>. Consequently, since the contribution of the granitic rocks to the total mass of most of the constructions is very low, the samples investigated could be used safely as building materials.

*Key words:* granitic rocks; natural radioactivity; building materials; dose assessment; Atticocycladic Zone

### Introduction

In igneous petrology, granite is a prevailing rock-type describing acid plutonic rocks having a particular mineralogy and geochemistry. However, in dimension stone market the term granite includes a variety of igneous and

metamorphic rock-types, used as building materials.

Granitic rocks (as a market term) are widely used as building and decorative materials due to their durability and appearance. Because of their mineralogical composition (e.g. zircon, monazite, apatite, titanite, xenotime), these rocks are likely

to contain high concentrations of natural radionuclides. Thus the study of the natural radioactivity in granitic rocks is an important subject in environmental radiological protection (Anjos et al., 2005) as it provides the possibility to assess any associated health hazard. Building materials contribute to natural radiation exposure in two ways: (a) by gamma radiation, from  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and their decay products, causing an external whole body exposure and (b) by radon exhalation causing an internal exposure due to deposition of radon decay products in the human respiratory tract. There are several studies regarding the measurement of natural radioactivity in granitic rocks used as building materials and the calculation of the associated dose rates, based on the concentration of radionuclides contained in those materials (Siotis and Wrixon, 1984; Savidou et al., 1995; Petropoulos et al., 2002; Anjos et al., 2005).

However, other than natural radioactivity, the physical, mechanical and technical characteristics of a rock, according to various national [American (ASTM), German (DIN), Italian (UNI), British (BS)] and international (ISO) standards, determine the suitability of a particular rock to be used as building material.

In the present contribution twenty three granite samples (petrologic term), collected from the Atticocycladic Zone are studied, aiming to show the suitability from radiological impact point of view, of these rocks for commercial use-building materials. Taking into account the natural radioactivity levels measured by using gamma-ray spectroscopy, the external exposure dose rate ( $D_a$ ) and hence the annual effective dose ( $H_E$ ) were estimated. Moreover, the external (gamma-ray) index as well as the contribution of the internal exposure due to radon inhalation are presented in order to consider the safe use of the specific granites as building materials. This study covers all possible granitic rock-types of a large part of Greece that belong to a unique geological zone, the Atticocycladic Zone. The basic purpose

of the present work aims at triggering the exploitation of Greek granites for the benefit of the national economy of the State, since the Atticocycladic Zone could be developed as the major provider of granite tiles to the most populated area of the country (Attica).

### Geological setting

The geology of the Atticocycladic Zone is characterized by numerous plutons of various compositions, which could be potentially used as building materials. The petrography, geochemistry, petrogenesis and age of the granites of this zone have been thoroughly studied by various researchers (Altherr et al., 1982; Altherr et al., 1988; Buick, 1991; Vekios, 1999; Pe-Piper et al., 1997; Pe-Piper, 2000; Altherr and Siebel, 2002; Pe-Piper and Piper, 2002; Pe-Piper et al., 2002; Mastrakas, 2006; Skarpelis et al., 2008; Iglseider et al., 2009; Stouraiti et al., 2010).

The Atticocycladic Zone is part of the Alpine orogen in the eastern Mediterranean and located between the Greek mainland and Anatolia. Published data on its plutonic rocks identify different types/provinces, based on differences in whole-rock chemistry, mineralogy, and isotopic composition and intrusion ages. Various plutons of Tertiary (Miocene-Oligocene) age intrude the metamorphic rocks of the Atticocycladic Zone. Most of the plutons are of granodiorite or granite composition and appear to be derived from crustal melts that mixed with mantle-derived mafic magma. They can be distinguished into: (a) I-type granites, granodiorites and dykes with similar composition, and (b) peraluminous two-mica granites and leucogranites with S-type characteristics.

### Materials and methods

Twenty three samples representing all major rock-types found in the granite occurrences of

the Atticocycladic Zone have been studied for their natural radioactivity. More specifically the samples were taken from Lavrio and from six Cycladic islands, namely the Serifos, Paros, Naxos, Mykonos, Delos, Tinos and Ikaria plutons (Figure 1). At this point it should be clarified that as long as Delos Island is an archeological reserve, it would not be a suitable source of building stones. However, samples from Delos Island were examined in order to provide data on the natural radioactivity of all the plutons of the Atticocycladic Zone.

A short description of each sample is given in Table 1. Further details on the mineralogical and chemical composition of the samples are given by Papadopoulos (2011). Their nomenclature is based on the Q-ANOR chemical classification (Streckeisen and Le Maitre, 1979).

Whole rock analyses were performed by ICP-ES in the ACME labs, Canada, using a SPECTRO CIROS VISION ICP-OES spectrometer. The samples were prepared by mixing with  $\text{LiBO}_2/\text{Li}_2\text{B}_4\text{O}_7$  flux. Crucibles were fused in a furnace. The cooled bead was dissolved in ACS grade nitric acid. Loss on ignition (LOI) was determined by igniting a sample split and then measuring the weight loss.

For the gamma-ray spectroscopy, the samples

were crushed into grains less than 400  $\mu\text{m}$  in diameter, oven-dried at 60 °C to constant weight, well-blended and measured using two high-resolution gamma ray spectrometry systems. The first one consisted of an HPGe coaxial detector with 42% efficiency and 2.0 keV resolution at 1.33 MeV photons, shielded by 4" Pb, 1mm Cd and 1mm Cu and the second one consisted of a LEGe planar detector with 0.7 keV resolution at 122 keV photons, shielded by 3.3" Fe-Pb, 1mm Cd and 1mm Cu.

The  $^{40}\text{K}$  content was obtained using its 1461 keV  $\gamma$ -ray. The  $^{232}\text{Th}$  content was calculated as the weighted mean value of  $^{228}\text{Ra}$  concentration (measured as  $^{228}\text{Ac}$ , using 911, 968 and 338 keV  $\gamma$ -rays) and  $^{228}\text{Th}$  concentration (measured as decay products in equilibrium, i.e.  $^{212}\text{Pb}$ , using 238 and 300 keV  $\gamma$ -rays,  $^{212}\text{Bi}$ , using 727 keV  $\gamma$ -ray and  $^{208}\text{Tl}$ , using 2614, 583 and 860 keV  $\gamma$ -rays). Likewise, the  $^{238}\text{U}$  content was calculated as the weighted mean value of  $^{234}\text{Th}$  concentration, using 63 keV  $\gamma$ -ray and  $^{234\text{m}}\text{Pa}$  concentration, using 776 and 1001 keV  $\gamma$ -rays. The efficiency calibration of the gamma spectrometry systems was performed with the radionuclide specific efficiency method in order to avoid any uncertainty in gamma ray intensities as well as the influence of coincidence summation

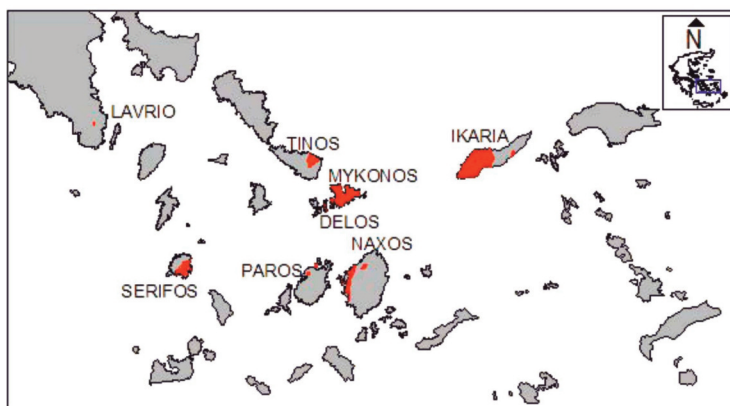


Figure 1. The plutonic rock occurrences of the Atticocycladic Zone.

Table 1. Petrographical features of the granite samples investigated.

| Origin  | Name | Color      | Grain size | Mineralogy                                     | Rock-type    |
|---------|------|------------|------------|--|--------------|
| Tinos   | FAL1 | Light grey | Medium     | Qz, Kf, Pl, Bi, Ap, Zr, Chl                    | GRT          |
|         | FAL2 | Grey       | Medium     | Qz, Kf, Pl, Bi, Hb, Chl, Tit, Zr, Ap           | bi GRD       |
| Mykonos | TUM1 | Light grey | Medium     | Qz, Kf, Pl, Hb, Bi, Ep, Tit, Thor, All, Zr, Ap | bi-hb GRT    |
|         | PLM1 | Light grey | Coarse     | Qz, Kf, Pl, Bi, Hb, Chl, Ap, Zr, Tit, Ep       | bi GRD       |
| Paros   | NP2  | Grey       | Fine       | Qz, Kf, Pl, Mu, Bi, Ap, Zr                     | GRT          |
|         | KP1  | Grey       | Fine       | Qz, Kf, Pl, Mu, Bi, Zr, Ap, Chl                | Two mica GRD |
| Naxos   | APN1 | Grey       | Coarse     | Qz, Kf, Pl, Bi, Hb, Ep, Ap, Zr                 | bi GRD       |
|         | AAN1 | Light grey | Medium     | Qz, Kf, Pl, Bi, Hb, Chl, Tit, Zr, Ep           | bi GRD       |
|         | KAN1 | White      | Fine       | Qz, Kf, Pl, Bi, Mu, Ap, Zr                     | GRT          |
|         | VN1  | Light grey | Coarse     | Qz, Kf, Pl, Hb, Bi, Chl, Ap, Zr                | hb GRT       |
| Ikaria  | API2 | Grey       | Coarse     | Qz, Kf, Pl, Zr, Ap, Thor, All, Xen             | GRT          |
|         | AI1  | Grey       | Coarse     | Qz, Kf, Pl, Bi, Chl, Ap, Zr                    | GRT          |
|         | KI2  | Grey       | Medium     | Qz, Kf, Pl, Bi, Ap, Zr                         | GRT          |
|         | MI1  | Light grey | Coarse     | Qz, Kf, Pl, Bi, Hb, Chl, Ap, Zr, Tit           | bi GRD       |
|         | PI1  | White      | Medium     | Qz, Kf, Pl, Bi, Mu, Zr, Ap, Mon, Xen           | GRT          |
|         | KI1  | Grey       | Medium     | Qz, Kf, Pl, Bi, Chl, Tit, Ap, Zr               | GRT          |
|         | XI3  | Grey       | Fine       | Qz, Kf, Pl, Mu, Bi, All, Zr, Ap                | GRT          |
| Serifos | KAS1 | Grey       | Medium     | Qz, Kf, Pl, Bi, Ap, Zr                         | bi GRD       |
|         | KS1  | Grey       | Medium     | Qz, Kf, Pl, Bi, Hb, Tit, All, Zr, Ap, Chl      | bi-hb GRT    |
|         | XS1  | Dark grey  | Medium     | Qz, Kf, Pl, Hb, Bi, Ap, Zr, Tit                | hb-bi GRD    |
| Lavrio  | L1   | Grey       | Fine       | Qz, Kf, Pl, Bi, Ap, Zr                         | bi GRD       |
| Delos   | DEL1 | Light grey | Medium     | Qz, Kf, Pl, Bi, Hb, Ap, Zr                     | bi GRD       |
|         | DEL2 | Grey       | Fine       | Qz, Kf, Pl, Bi, Hb, Ap, Zr                     | GRT          |

Qz = quartz; Kf = K-feldspars; Pl = plagioclase; Bi(bi) = biotite; Hb(hb) = hornblende; Mu = muscovite; Ap = apatite; Zr = zircon; Ep = epidote; All = allanite; Tit = titanite; Chl = chlorite; Thor = thorite; Xen = xenotime; GRT = granite; GRD = granodiorite.

and self-absorption effects of the emitting gamma photons. A set of high quality certified reference materials (IAEA, RGU-1, RGTh-1, RGK-1) (I.A.E.A., 1987) was used, with densities similar to the building materials measured after pulverization. Cylindrical and Marinelli geometry were used assuming that the radioactivity is homogeneously distributed in the measuring samples. The measurement duration was up to

200000 s. The total uncertainty of the radioactivity levels was calculated by propagation of the systematic and random errors of measurements. The systematic errors in the efficiency calibration ranges from 0.5-2% and the random errors of the radioactivity measurements extend up to 5%, except in the  $^{238}\text{U}$  measurement, where the error extends up to 12%. Finally, the minimum detectable specific activities for  $^{238}\text{U}$ ,

$^{232}\text{Th}$  and  $^{40}\text{K}$  is 1.6, 9 and 26 Bq kg<sup>-1</sup> respectively (Stoulos et al., 2003).

#### *Derivation of the dose rates*

Aiming to protect the public from excessive exposure to radioactivity, various radioactivity indices have been proposed in order to assess the exposure to natural radioactivity of building materials.

(1) United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The indices determine the absorbed dose rate and the annual effective dose (UNSCEAR, 1993).

a) Absorbed gamma dose rate (Da). This index calculates the energy transfer rate of an ionizing radiation which is absorbed per unit mass in a medium. In order to facilitate the calculation of dose rates in air from different combinations of three radionuclides in building materials and by applying the appropriate conversion factors, an activity utilization index is constructed that is given by the following expression:

$$[(C_{\text{Th}}/A_{\text{Th}})*f_{\text{Th}} + (C_{\text{U}}/A_{\text{U}})*f_{\text{U}} + (C_{\text{K}}/A_{\text{K}})*f_{\text{K}}]*w_{\text{m}} \quad (1)$$

where  $C_{\text{U}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  are the specific activity concentrations (Bq kg<sup>-1</sup>) of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the building materials considered;  $f_{\text{Th}}$ ,  $f_{\text{U}}$  and  $f_{\text{K}}$  are the fractional contributions to the total dose rate in air due to gamma radiation from the actual concentrations of these radionuclides.  $A_{\text{Th}}$ ,  $A_{\text{U}}$  and  $A_{\text{K}}$  are the typical activities per unit mass of  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and  $^{40}\text{K}$ , which are equal to 50, 50 and 500 Bq kg<sup>-1</sup> (N.E.A., 1979). The activity utilization index is multiplied by a factor ( $w_{\text{m}}$ ) that represents the fractional usage of those materials in the dwelling with the characteristic activity. Full mass utilization ( $w_{\text{m}} = 1$ ) of a given material, implies that all building materials used in a model masonry house are composed of this specific material. Half mass utilization ( $w_{\text{m}} = 0.5$ ) means that 50% of the masonry mass is composed of the material considered, and so on. For full mass

utilization of a model masonry house ( $C_{\text{Th}} = C_{\text{U}} = 50$  Bq kg<sup>-1</sup> and  $C_{\text{K}} = 500$  Bq kg<sup>-1</sup>), the activity utilization index is unity by definition and is deemed to imply a dose rate of 80 nGy h<sup>-1</sup> (UNSCEAR, 1993).

b) Annual effective dose (HE). This index determines the biological impact of the exposure to radiation. The effective dose rate due to external exposure in mSv y<sup>-1</sup>, is calculated by the following formula:

$$H_{\text{Ext}} = 10^{-6} \times D \times T \times F \quad (2)$$

where  $D$  is the calculated dose rate in nGy h<sup>-1</sup>,  $T$  is the indoor occupancy time, which implies that 20% of time is spent outdoors, and is equal to 7000 h/y, and  $F$  is the dose conversion factor equal to 0.7 Sv/Gy. The  $H_{\text{Ext}}$  should be less than 1 mSv y<sup>-1</sup> (UNSCEAR, 1993; 2000).

(2) European Commission. It has proposed the Gamma-ray index ( $I_{\gamma}$ ), which is strongly related to the annual effective dose (E.C. 1999). The gamma-ray index is calculated considering a standard room model with dimensions of 4 m x 5 m x 2.8 m and 20 cm thick walls. This index factor associated to the external exposure is calculated by the formula:

$$I_{\gamma} = C_{\text{U}} / 300 \text{ Bq kg}^{-1} + C_{\text{Th}} / 200 \text{ Bq kg}^{-1} + C_{\text{K}} / 3000 \text{ Bq kg}^{-1} \quad (3)$$

and is correlated with the annual dose rate due to gamma radiation (external exposure). Materials having  $I_{\gamma} < 2$  would increase the annual effective dose by 0.3 mSv, while for  $2 < I_{\gamma} < 6$ , the gamma-ray index corresponds to an increase in effective dose by 1 mSv y<sup>-1</sup>. Building materials used superficially rather than in bulk amounts (tiles, boards, etc.) should be exempted from all restrictions concerning radioactivity, if the excess of gamma radiation originating from them increases the annual effective dose of a member of public by 0.3 mSv at the most. On the other hand, dose rates higher than 1 mSv y<sup>-1</sup> is allowed only

in exceptional cases, where materials are locally used. Finally, samples with  $I_\gamma > 6$  cannot be recommended for use in buildings (E.C., 1999).

### Results and discussion

The specific activities ( $\text{Bq kg}^{-1}$ ) of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  of the samples, along with some statistical parameters are given in Table 2. The sample displaying the highest specific activity of  $^{238}\text{U}$  ( $229 \pm 10$ ) is API-2 from Ikaria, followed by samples TUM-1 (Mykonos) and PI-1 (Ikaria) with specific activities of  $^{238}\text{U}$   $196 \pm 6$  and  $179 \pm 9$   $\text{Bq kg}^{-1}$ , respectively. Moreover, eight (8) of the

studied samples exceed the average specific activity of  $^{238}\text{U}$  of granites which is  $78 \text{ Bq kg}^{-1}$  (Table 3) (UNSCEAR, 2000). The samples having the highest specific activities of  $^{232}\text{Th}$  are VN-1 and APN-1 from Naxos ( $132 \pm 2$  and  $105 \pm 3$   $\text{Bq kg}^{-1}$ , respectively), followed by TUM-1 (Mykonos) and DEL-1 (Delos) with  $99 \pm 2$   $\text{Bq kg}^{-1}$  for both of them. Comparing the specific activities of  $^{232}\text{Th}$  of the samples with the average granite, it appears that only one sample (VN-1) exceeds the value of  $111 \text{ Bq kg}^{-1}$  (UNSCEAR, 2000). As far as the specific activities of  $^{40}\text{K}$  are concerned, the samples DEL-2 (Delos), PI-1 (Ikaria) and VN-1 (Naxos) display the highest

Table 2. Specific activities ( $\text{Bq kg}^{-1}$ ) of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  for the granite samples examined.

| Name          | Activity concentrations ( $\text{Bq kg}^{-1}$ ) |             |                   |             |                 |             |
|---------------|---|-------------|-------------------|-------------|-----------------|-------------|
|               | $^{238}\text{U}$                                | $\pm\sigma$ | $^{232}\text{Th}$ | $\pm\sigma$ | $^{40}\text{K}$ | $\pm\sigma$ |
| FAL-1         | 71.0  | 6.0         | 67.0              | 2.0         | 1021.0          | 14.0        |
| FAL-2         | 73.0  | 7.0         | 62.0              | 2.0         | 990.0           | 14.0        |
| TUM-1         | 196.0   | 6.0         | 99.0              | 2.0         | 1094.0          | 16.0        |
| PLM-1         | 72.0  | 7.0         | 83.0              | 1.0         | 853.0           | 12.0        |
| NP-2          | 57.0  | 7.0         | 65.0              | 2.0         | 1095.0          | 16.0        |
| KP-1          | 27.0  | 6.0         | 57.0              | 2.0         | 798.0           | 12.0        |
| APN-1         | 81.0  | 8.0         | 105.0             | 3.0         | 996.0           | 14.0        |
| AAN-1         | 87.0  | 9.0         | 86.0              | 2.0         | 962.0           | 15.0        |
| KAN-1         | 31.0  | 6.0         | 72.0              | 2.0         | 1133.0          | 16.0        |
| VN-1          | 99.0  | 8.0         | 132.0             | 2.0         | 1141.0          | 17.0        |
| API-2         | 229.0   | 10.0        | 96.0              | 2.0         | 1064.0          | 12.0        |
| AI-1          | 85.0  | 8.0         | 97.0              | 3.0         | 1138.0          | 15.0        |
| KI-2          | 94.2  | 8.7         | 87.0              | 3.0         | 1046.0          | 16.0        |
| MI-1          | 72.0  | 8.0         | 90.0              | 2.0         | 941.0           | 13.0        |
| PI-1          | 179.0   | 9.0         | 65.0              | 2.0         | 1145.0          | 16.0        |
| KI-1          | 67.0  | 8.0         | 81.0              | 2.0         | 1055.0          | 14.0        |
| XI-3          | 71.0  | 7.0         | 41.0              | 1.0         | 974.0           | 15.0        |
| KAS-1         | 48.0  | 7.0         | 46.0              | 1.0         | 807.0           | 10.0        |
| KS-1          | 59.7  | 7.6         | 52.3              | 1.7         | 873.6           | 13.2        |
| XS-1          | 67.0  | 5.0         | 46.0              | 1.0         | 761.0           | 15.0        |
| L-1           | 66.0  | 7.0         | 57.0              | 2.0         | 788.0           | 12.0        |
| DEL-1         | 56.0  | 8.0         | 99.0              | 2.0         | 851.0           | 13.0        |
| DEL-2         | 44.0  | 7.0         | 80.0              | 2.0         | 1220.0          | 17.0        |
| Average       | 83.5  |             | 76.8              |             | 989.0           |             |
| Range         | 27.0-229.0                                      |             | 41.0-132.0        |             | 761.0-1220.0    |             |
| St. Deviation | 51.4  |             | 22.6              |             | 134.2           |             |

Table 3. Specific activities (Bq kg<sup>-1</sup>) of <sup>238</sup>U and <sup>232</sup>Th of various rocks (UNSCEAR, 1993).

| Rock type       | <sup>238</sup> U |         | <sup>232</sup> Th |          |
|-----------------|------------------|---------|-------------------|----------|
|                 | Average          | Range   | Average           | Range    |
| Granite         | 78               | 1-370   | 111               | 0.4-1030 |
| Basalt          | 11               | 0.4-41  | 10                | 0.2-3.6  |
| Limestone       | 45               | 0.4-340 | 60                | 0.1-540  |
| Sandstone/Shale | 60               | 1-990   | 50                | 0.8-1470 |
| Gneiss          | 50               | 1-1800  | 60                | 0.4-420  |
| Schist          | 37               | 1-660   | 49                | 0.4-370  |

values (1220±17, 1145±16 and 1141±17 Bq kg<sup>-1</sup>). These samples, along with KAN-1 (Naxos), AI-1 (Ikaria) exceed the average granite value for <sup>40</sup>K, which is 1104 Bq kg<sup>-1</sup> (UNSCEAR, 2000).

The specific activities of the granites of the Atticocycladic Zone (AZG) were compared to the respective values given by the commercial granitic rocks imported in Greece (IGG) (Pavlidou et al., 2006) and Cyprus (ICG) (Tzortzis et al., 2003) (Table 4). The average values and the standard deviations of the AZG are 83.5±51.4, 76.8±22.6 and 989.0±134.2 for <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, respectively. The average specific activities (Bq kg<sup>-1</sup>) of the sixteen (16) granitic samples imported in Greece and the twenty-eight 28 samples of the granitic samples imported in Cyprus are 64±53 (<sup>238</sup>U), 81±79 (<sup>232</sup>Th) and 1104±407 (<sup>40</sup>K) and 77±116 (<sup>238</sup>U), 180±143 (<sup>232</sup>Th) and 1215±352 (<sup>40</sup>K), respectively. It is evident that the specific activities of <sup>232</sup>Th and <sup>40</sup>K are higher in the IGG and ICG, compared to the

AZG. Finally, the AZG, IGG and ICG appear to have similar specific activities of <sup>238</sup>U.

The values of the radioactivity indices of the AZG are given in Table 5. The external absorbed dose rate and so the effective dose rate were calculated assuming that the room has been constructed totally by granite. The samples with the higher radioactivity indices in all cases are API-2 (Ikaria), VN-1 (Naxos), PI-1 (Ikaria), AI-1 (Ikaria), APN-1 (Naxos) and TUM-1 (Mykonos). The gamma-ray index values ( $I_\gamma < 2$ ) of the samples, imply that all of them can be used as building materials without any restrictions (Figure 2).

Uranium series contributes also to an internal exposure due to radon inhalation. The average annual effective dose to lungs due to radon is proportional to radon exhalation from building materials. Measurements on radon exhalation from typical Greek building materials demonstrate that granite tiles have lower

Table 4. Specific activities (Bq kg<sup>-1</sup>) of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K of the granitic rocks imported in Greece (Pavlidou et al., 2006) and Cyprus (Tzortzis et al., 2003).

| Radionuclide       | IGG              |                   |                 | ICG              |                   |                 |
|--------------------|------------------|-------------------|-----------------|------------------|-------------------|-----------------|
|                    | <sup>238</sup> U | <sup>232</sup> Th | <sup>40</sup> K | <sup>238</sup> U | <sup>232</sup> Th | <sup>40</sup> K |
| Average            | 63.7             | 81                | 1104            | 77.2             | 143.4             | 1215            |
| Range              | 1.6-170          | <DL*-354          | 49-1592         | 1-588            | 0-906             | 50-1606         |
| Standard Deviation | 53               | 79                | 407             | 116              | 180               | 352             |

\*DL = Detection Limit

Table 5. Absorbed gamma dose rate  $D_a$ , Annual effective dose  $H_E$  (mSv) and gamma ray index  $I_\gamma$  for the granite samples examined.

| Name          | $D_a$ (nGy/h) | $H_E$ (mSv/y) | $I_\gamma$ |
|---------------|---------------|---------------|------------|
| FAL-1         | 120.8         | 0.6           | 0.9        |
| FAL-2         | 124.7         | 0.6           | 1.0        |
| TUM-1         | 140.9         | 0.7           | 1.1        |
| PLM-1         | 124.2         | 0.6           | 1.0        |
| NP-2          | 111.6         | 0.6           | 0.9        |
| KP-1          | 80.8          | 0.4           | 0.6        |
| APN-1         | 144.2         | 0.7           | 1.1        |
| AAN-1         | 128.4         | 0.6           | 1.0        |
| KAN-1         | 106.8         | 0.5           | 0.8        |
| VN-1          | 175.6         | 0.9           | 1.4        |
| API-2         | 198.8         | 1.0           | 1.5        |
| AI-1          | 156.4         | 0.8           | 1.2        |
| KI-2          | 138.0         | 0.7           | 1.1        |
| MI-1          | 126.0         | 0.6           | 1.0        |
| PI-1          | 172.1         | 0.8           | 1.3        |
| KI-1          | 128.1         | 0.6           | 1.0        |
| XI-3          | 104.0         | 0.5           | 0.8        |
| KAS-1         | 84.7          | 0.4           | 0.7        |
| KS-1          | 92.0          | 0.5           | 0.7        |
| XS-1          | 90.0          | 0.4           | 0.7        |
| L-1           | 96.7          | 0.5           | 0.8        |
| DEL-1         | 121.6         | 0.6           | 1.0        |
| DEL-2         | 119.5         | 0.6           | 0.9        |
| Range         | 80.8-198.8    | 0.4-1.0       | 0.6-1.5    |
| Average       | 125.5         | 0.6           | 1.0        |
| St. Deviation | 29.9          | 0.2           | 0.2        |

exhalation rate than concrete. Even some granite tiles with extremely high natural radioactivity, exhibit normal exhalation rates, because of their limited usage (relative to concrete) in dwellings. So, negligible influence on internal exposure due to granite tiles is expected since concrete is the major contributor to internal exposure (Stoulos et al., 2003). Assuming that the standard room has been fully constructed of granite, the average annual effective dose rate to lungs ranges between 0.04-0.4 mSv. These values are lower than the effective dose rate from external exposure, which ranges between 0.4-1 mSv  $y^{-1}$ . The total effective dose rate received indoors from granites should be expected between 0.4 and 1.4 mSv  $y^{-1}$ . However, for a building material like granite, Chen and Lin (1996) proposing a different standard room model of 6 m x 4 m x 3 m dimensions with 0.2 m thick wall and 0.02 m thick granite floor as a more reasonable model, estimated that the granite material in this construction was only 2.2% of the total weight.

The radioactivity indices of AZG are compared to the average values of both the IGG and ICG (Table 6 and Figure 3). The absorbed dose rate, the annual effective dose and the gamma-ray index of the AZG is  $125.5 \pm 29.9$  nGy  $h^{-1}$ ,  $0.6 \pm 0.2$  and  $1.0 \pm 0.2$ , respectively. The majority of the AZG samples exhibit lower radioactivity indices

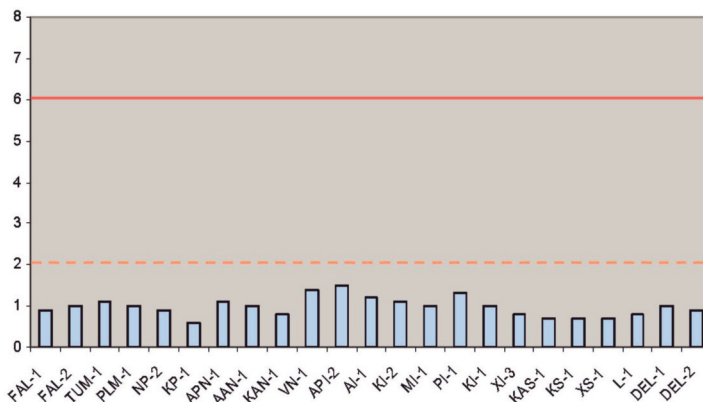


Figure 2. Gamma-ray index values of the samples in comparison to the limits.



Table 6. Values of Absorbed Dose rate, Annual Effective Dose and Gamma-Ray index of the granitic rocks imported in Greece (Pavlidou et al., 2006) and Cyprus (Tzortzis et al., 2003).

|                    | IGG   |   |   | ICG   |                                      |   |
|--------------------|---|---|---|---|--------------------------------------|---|
|                    | D <sub>a</sub><br>Absorbed<br>Dose rate<br>(nGy h <sup>-1</sup> ) | I <sub>γ</sub><br>Gamma<br>ray<br>Index | H <sub>Ext</sub><br>Annual<br>Effective<br>Dose<br>(mSv y <sup>-1</sup> ) | D <sub>a</sub><br>Absorbed<br>Dose rate<br>(nGy h <sup>-1</sup> ) | I <sub>γ</sub><br>Gamma<br>ray Index | H <sub>Ext</sub><br>Annual<br>Effective<br>Dose<br>(mSv y <sup>-1</sup> ) |
| Average            | 125.4   | 1.0                                     | 0.6   | 124.9   | 1.0                                  | 0.6   |
| Range              | 8.4-364.8   | 0.1-2.9                                 | 0.01-1.8  | 0.5-835.5   | 0.0-6.5                              | 0.0-4.1   |
| Standard Deviation | 79.9  | 0.6                                     | 0.4   | 159.2   | 1.2                                  | 0.8   |

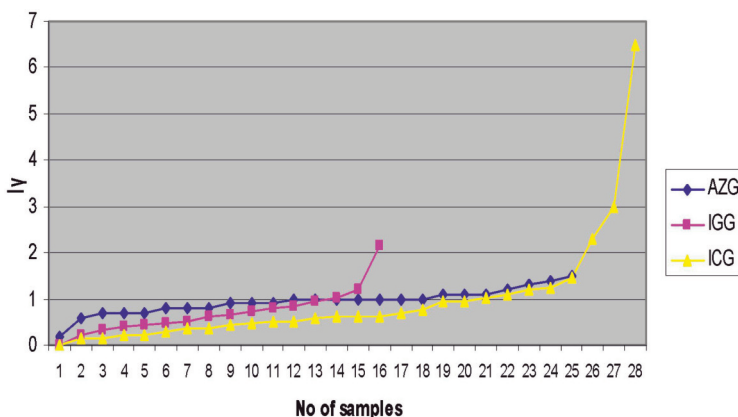


Figure 3. Gamma-ray index values of AZG, IGG and ICG.

than the IGG and ICG samples. Moreover, the maximum values of these indices in the IGG and ICG is by far higher than those of AZG. The relatively higher standard deviation displayed for all radioactivity indices in the case of IGG and ICG can be explained by the fact that these rocks are “granites” only in the commercial sense of the term. In fact they are igneous rocks, ranging from gabbros to granites. The AZG rocks are more uniform in composition, as all of them are felsic igneous plutonic rocks (granites and granodiorites).

The mineral composition of an igneous rock also plays a key role to its content in U and Th. According to Faure (1986), U- and Th-rich

minerals are much more abundant in felsic igneous rocks than in mafic ones. This can be explained by the incompatibility of both U and Th during partial melting and fractional crystallization processes ( $K_d < 1$ ), leading thus in the remaining of U and Th in the melt and their incorporation in minerals of acid rocks. The above must also be taken into account, as the specific activity concentration in natural radionuclides and the radioactivity indices of the AZG is similar or lower than the IGG and ICG. Consequently, if all the samples of mafic and intermediate composition are excluded, the natural radioactivity of AZG would be by far lower than that of the commercial types.

## Conclusions

Exploitation of high-resolution  $\gamma$ -ray spectroscopy provides a sensitive experimental tool in studying natural radioactivity in various rock types. Some of the studied granitic samples reveal elevated specific activities and elemental concentrations of Th, U and K, thus contributing higher absorbed dose rates in air comparing to other building materials.

The granitic rocks of the Atticocycladic Zone display similar specific activity of  $^{238}\text{U}$  and lower specific activity of  $^{232}\text{Th}$  to those of the average granite. The absorbed, the annual effective dose and the gamma-ray index were determined, so as to assess the radiological impact from the granites investigated, in case they were used as building materials. The absorbed dose rate due to external radiation ranges around a mean value of  $86 \pm 30 \text{ nGy h}^{-1}$  and the annual effective dose and the gamma index around  $0.6 \pm 0.2 \text{ mSv y}^{-1}$  and  $1.0 \pm 0.2$ , respectively. Taking into consideration the internal exposure due to radon inhalation and that the room is fully constructed by granite the total annual effective dose ranges between 0.4 and  $1.4 \text{ mSv y}^{-1}$ . Moreover, all of the samples meet the exemption dose limit of  $0.3 \text{ mSv y}^{-1}$ , as proposed by the European Commission. Therefore, at least from radiological point of view and for the investigated samples, the use of granites from the Atticocycladic zone as building materials is recommended. The average values of the above indices are lower than those of the imported in Greece and Cyprus granitic rocks. Considering the benefits for the national economy of Greece, the possibility of exploiting these rocks should be investigated more thoroughly by the mining industry.

## Acknowledgements

The authors are grateful to Georgia Pe-Piper and an anonymous referee for their valuable comments that improved the manuscript.

## References

- Anjos R.M., Veiga R., Soares T., Santos A.M.A., Aguiar J.G., Frasca M.H.B.O., Brage J.A.P., Uzeda D., Mangia L., Facure A., Mosquera B., Carvalho C. and Gomes P.R.S. (2005). Natural radionuclide distribution in Brazilian commercial granites. *Radiation Measurements*, 39, 245-253.
- Altherr R., Kreuzer H., Wendt, I. Lenz, H., Wagner G.A., Keller J., Harre W. and Hohndorf A. (1982). A late Oligocene/early Miocene high temperature belt in the Attic-Cycladic crystalline complex (SE Pelagonian, Greece). *Geologisches Jahrbuch*, E23, 97-164.
- Altherr R., Henjes-Kunst F.J., Matthews A., Friedrichsen H. and Hansen B.T. (1988). O-Sr isotopic variations in Miocene granitoids from the Aegean: evidence for an origin by combined assimilation and fractional crystallisation. *Contributions to Mineralogy and Petrology*, 100, 528-541.
- Altherr R. and Siebel W. (2002). I-type plutonism in a continental back arc setting: Miocene granitoids and monzonites from the central Aegean Sea, Greece. *Contributions to Mineralogy and Petrology*, 143, 397-415.
- Buick I.S. (1991). The late Alpine evolution of an extensional shear zone, Naxos, Greece. *Journal of the Geological Society of London*, 148, 93-103.
- Chen C.J. and Lin Y.M. (1996). Assessment of building materials for compliance with regulations of ROC. *Environment International*, 22, 221-226.
- European Commission (E.C.) (1999). Radiation Protection 112 : Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials Directorate General Environment, Nuclear Safety and Civil Protection.
- Faure G. (1986). Principles of Isotope Geology, second ed. John Wiley & Sons, London.
- I.A.E.A. (International Atomic Energy Agency) (1987). Preparation of Gamma-ray Spectroscopy Reference Materials RGU-1, RGTh-1 and RGK-1 Report-IAEA/RL/148, Vienna (1987).
- Iglseder C., Grasemann B., Schneider D.A., Petrakakis K., Miller C., Klötzli U.S., Thöni M., Zámolyi A. and Rambousek C. (2009). I and S-type plutonism on Serifos (W-Cyclades, Greece). *Tectonophysics*, 473, 69-83.
- Mastrakas N. (2006). Tinos pluton and associated

- skarn formations. PhD thesis, Univ. Patras, 227 pp.
- Nuclear Energy Agency (N.E.A.), (1979). Exposure to radiation from the natural radioactivity in building materials. Report by NEA Group of Experts, OECD, Paris.
- Papadopoulos A. (2011). Natural radioactivity in relation to mineralogy, geochemistry of uranium and thorium of magmatic rocks from Greece: Contribution to the use of natural building materials. PhD Thesis, Aristotle University of Thessaloniki.
- 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.
- Pe-Piper G., Kotopouli C.N. and Piper D.J.W. (1997). Granitoid rocks of Naxos, Greece: regional geology and petrology. *Geological Journal*, 32, 153-171.
- Pe-Piper G. (2000). Origin of S-type granites coeval with I-type granites in the Hellenic subduction system, Miocene of Naxos, Greece. *European Journal of Mineralogy*, 12, 859-875.
- Pe-Piper G. and Piper D.J.W. (2002). The igneous rocks of Greece: the anatomy of an orogen, Borntraeger, Berlin.
- Pe-Piper G., Piper D.J.W. and Matarangas D. (2002). Regional implications of geochemistry and style of emplacement of Miocene I-type diorite and granite, Delos, Cyclades, Greece. *Lithos*, 60, 47-66.
- Petropoulos N.P., Anagnostakis M.J., and Simopoulos S.E. (2002). Photon attenuation, natural radioactivity content and radon exhalation rate of building materials. *Journal of Environmental Radioactivity*, 61, 257-269.
- Savidou A., Raptis C. and Kritidis P. (1995). Natural radioactivity and radon exhalation from building materials used in Attica region, Greece. *Radiation Protection Dosimetry*, 59, 309-312.
- Siotis I. and Wrixon A.D. (1984). Radiological consequences of the use of fly ash in building materials in Greece. *Radiation Protection Dosimetry*, 7, 101-105.
- Skarpelis N., Tsikouras B. and Pe-Piper G. (2008). The Miocene igneous rocks in the Basal Unit of Lavrion (SE Attica, Greece): petrology and geodynamic implications. *Geological Magazine* 145, 1-15.
- Stouraiti C., Mitropoulos P., Tarney J., Barreiro B., McGrath A.M. and Baltatzis E. (2010). Geochemistry and petrogenesis of late Miocene granitoids, Cyclades, southern Aegean: Nature of source components. *Lithos*, 114, 337-352.
- 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.
- Streckeisen A. and Le Maitre R.W. (1979). A chemical approximation to the modal QAPF classification of the igneous rocks. *Neues Jahrbuch für Mineralogie Abh.*, 136, 169-206.
- Tzortzis M., Tsertos H., Christofides S. and Christodoulides G. (2003). Gamma measurements and dose rates in commercially used tiling rocks (granites). *Journal of Environmental Radioactivity*, 70, 223-235.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (1993). Sources and Effects of Ionising Radiation Vol. I. United Nations, New York.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000). Sources and Effects of Ionising Radiation Vol. I. United Nations, New York.
- Vekios P. (1999). Sedimentological and geochemical study of silcrete formation of the island of Ikaria and the related uraniferous mineralization. PhD thesis. University of Athens.

Submitted, April 2012 - Accepted, November 2012

