# Natural radioactivity distribution and gamma radiation exposure of beach sands close the granitoids of NE Chalkidiki, Greece

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Abstract
This study aims to evaluate the activity concentrations of $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{228}Th$ and $^{40}K$ along the beaches NE Chalkidiki being adjacent to the local granitoids. These ranged from 9.0-63.0, 11.5-62.9, 10.9-54.5, 11.8-56.6 and 27-828 Bq/kg respectively. The (% wt.) heavy magnetic (HM) (allanite, epidote, amphibole, mica, clinopyroxene, magnetite and pyrite) fraction, the heavy non-magnetic (HNM) (zircon, titanite and apatite) fraction and the total heavy fraction (TH), were correlated with the concentrations of the measured radionuclides in the bulk samples. The measured radionuclides in the beach sands were normalized to the respective values measured in the granitic rocks, which are their most probable parental rocks, so as to provide data upon their enrichment or depletion. The annual effective dose varies between 0.003 and 0.009 mSv $y^{-1}$ for tourists and from 0.012 to 0.037 mSv $y^{-1}$ for local people working on the beach.

Keywords: natural radioactivity, beach sands, granitic rocks, heavy magnetic and non-magnetic fractions, annual effective dose equivalent

## 1. Introduction

Beach sands are composed mainly of quartz, feldspar and other minerals resistant to wave abrasion. They are the products of a combination of weathering, fragmentation, and degradation (Pettijohn et al. 1987). Beach placer or “black sand” deposits around the world are known for their economic concentrations of heavy minerals such as monazite, zircon, ilmenite, rutile, garnet, allanite and sillimanite (Alam et al. 1999; Freitas and Alencar 2004).
Studies concerning the radiation hazards arising from the use of sand or soil have shown that natural radiation is the largest contributor to external dose to the world population. The study of the distribution of natural radionuclides ($^{238}U$, $^{232}Th$, their daughter products and $^{40}K$) allows the understanding of the radiological implication of these elements due to the gamma-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters. Therefore, the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population (UNSCEAR 1993, 2000). Various studies on natural radioactivity levels of sands have been carried out worldwide concerning in particular areas with high background radiation like India and Brazil or countries with extended sand landscapes. Exposure dose rates of the public have been also assessed indicating that these dose rates vary depending upon the concentration of the natural radionuclides present in sands and bed-rocks, which in turn depend upon the local geology of each region (Hammoud 1966, De Meijer et al. 1988, 2001, Dabbour 1995, Alam et al. 1999, Kannan et al. 2002, Freitas and Alencar 2004, Mahmoud et al. 2004, Mohanty et al. 2004, Singh et al. 2005, Veiga et al. 2006, Vassas et al. 2006, Harb 2008, Shetty et al. 2011, Nada et al. 2012).
The coastline of Greece is one of the largest worldwide; however, despite of the fact that most of Greek beaches are highly touristic, there is no data upon their natural radioactivity levels until now. However, heavy minerals-rich beach sands (black sands) which are usually associated with high levels of natural radioactivity have been reported in Greece, more

specifically in Sithonia (Papadopoulos et al. 2014), in Touzla area near Thessaloniki (Filippidis et al. 1997) and in N. Peramos near Kavala (Pergamalis et al. 2001).

The beaches close to Chalkidiki, are one of the most touristic areas of Northern Greece. Close to the beaches of NE Chalkidiki, the granitic rocks of Ierissos-Ouranoupoli and Stratoni plutons are present, which are well known for their elevated natural radioactivity levels compared to other natural stones. This study is focused on the beach sands that are placed adjacent to the rock-types of the granitoids of NE Chalkidiki, which are their most probable parental rock. Any existing data on the natural radioactivity of these rocks are used.

The main goal of this work is to assess the activity concentrations of the natural radionuclides and the radioactivity indices related to them. Any relation of several mineral fractions separated from the whole sample (total heavy, heavy magnetic and heavy non-magnetic % wt.) and mineral constituents is also assessed. Correlations between the REE content and the natural radionuclides are also assessed. The evaluation of the enrichment or depletion of the natural radionuclides in beach sands relative to the granitic rocks (parental rocks) is also attempted. Moreover, the external gamma index, the absorbed gamma dose rate and the annual effective dose received by the population due to their exposure on gamma radiation by the beach sands is also presented.

## 2. Materials and methods

### 2.1 Geological Setting

Both Ierissos-Ouranoupoli and Stratoni plutons intrude the Sebomacedonian Massif and more specifically the Kerdylion Unit. The latter is mainly composed of mica and amphibole gneisses, marbles and amphibolites and belong to the pre-Alpine metamorphic basement (Kilias 1999, Kydonakis et al. 2014 and references therein).

#### 2.1.1 Ierissos-Ouranoupoli pluton

The main rock-type present is biotite granodiorite. Its constituent minerals are quartz, feldspars, biotite, epidote, magnetite, apatite, zircon, allanite and titanite. The rock-types of Ierissos-Ouranoupoli pluton are high-K calc-alkaline to shoshonitic (Perugini et al. 2004). The age of the pluton has been determined at 54 Ma using U-Pb dating on uraninite (Frei 1992).

#### 2.1.2 Stratoni pluton

The major rock-type present is biotite granodiorite. The constituent minerals are quartz, feldspars, biotite, hornblende, titanite, apatite, magnetite, epidote, chlorite and sericite. The rock-types of this pluton are calc-alkaline to high-K calc-alkaline (Perugini et al. 2004). According to Papadakis (1971), the age of the pluton is 29.6 Ma, using K-Ar dating on biotite.

### 2.2 Collection and pre-treatment of the samples

Representative sediment samples obtained from 6 random points (separated each other by around 500m) were collected from beaches, close to the granitoid rocks of NE Chalkidiki (Fig. 1, Tab. 1), using sampling intervals relative to the length of each beach. One sample (SSTR1C) was collected from a specific horizon of dark brown color and thickness of 1-4 cm, heavily enriched in heavy minerals, especially pyrite (Tab. 1, Fig. 2).

For each sand sample analyzed, three sub-samples of equal mass (≈500g) were obtained from a depth of 20 cm. The sub-samples, corresponded to 3 different points, forming an equilateral triangle (with dimensions of ≈1.4 m) corresponding to an area of approximately 1 $m^2$. The three sub-samples were homogenized by mixing in situ and this sand mixture, weighing approximately 1.5 kg, was considered as representative.

In the laboratory, the samples were cleaned with warm water and dried. Any coarse wastes (sea shells, etc.) were removed during sieving. The 8, 4, 2, 1, 0.5, 0.125 and 0.063 mm sieves were used to obtain the grain-size distribution of the samples. For the mineral separations, the 0.125-0.5 mm grain-size fraction was used, after the determination of the average grain size of the heavy minerals under the binocular microscope. After magnetite removal using a hand magnet, heavy liquid (tetrabromoethane, 2.967 g/cm$^3$) and a Frantz isodynamic separator were employed to determine the wt % heavy fraction and the heavy magnetic and non-magnetic fractions of the whole sample. The heavy magnetic fraction (<0.8 amp at forward and side slope of 15° and 25°, respectively) contains amphibole, mica, magnetite and pyrite while the heavy non-magnetic fraction (>0.8 amp at same settings) contains zircon, titanite and apatite. All the above mentioned minerals have been identified under the binocular microscope. Sample preparation and mineral separations were performed at the laboratories of the Department of Mineralogy-Petrology-Economic Geology, School of Geology; Aristotle University of Thessaloniki.

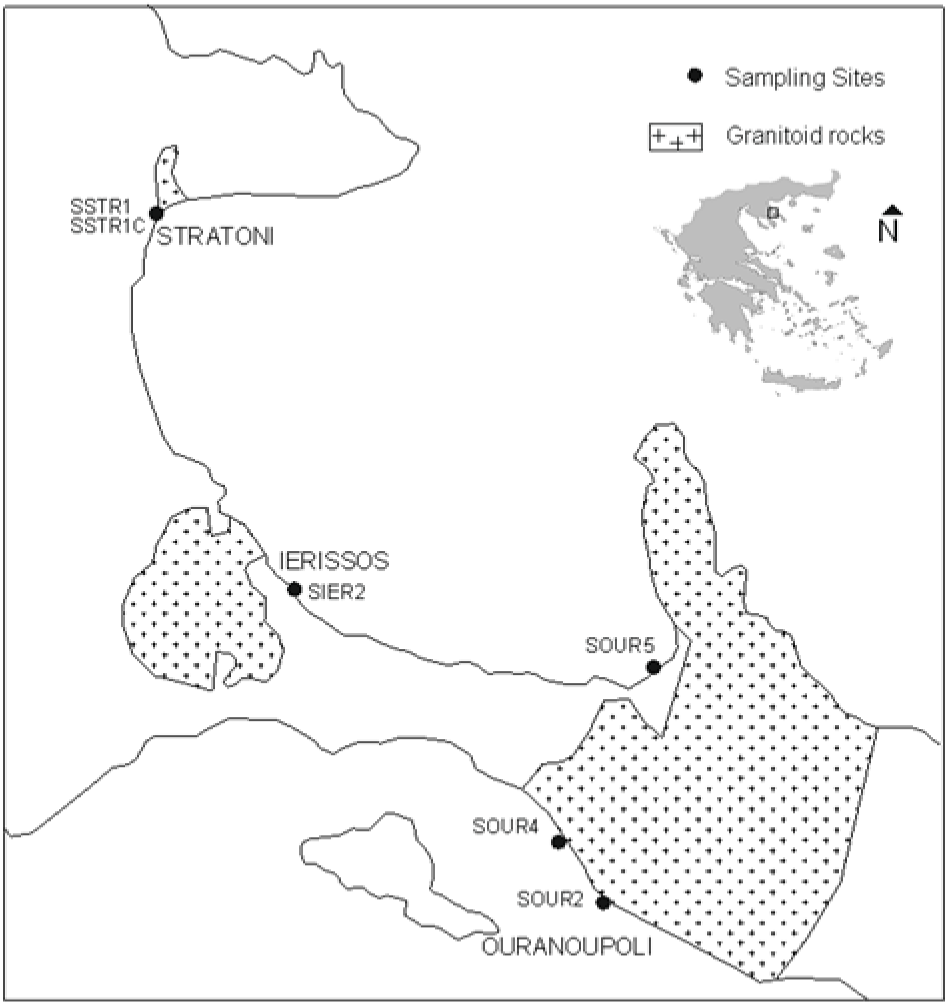


**Figure 1.** Geological sketch NE Chalkidiki presenting the sampling sites.

**Table 1.** Location, neighboring granitic rocks sample and textural characteristics of the samples studied.

| Sample | Location (beach name) | Neighboring granitoid rock sample | Comments | SAMPLE TYPE | TEXTURAL GROUP | SEDIMENT NAME |
|---|---|---|---|---|---|---|
| SSTR1C | Stratoni | STR1 | Heavy minerals horizon | Unimodal, Very Well Sorted | Sand | Very Well Sorted Fine Sand |
| SSTR1A | | | | Trimodal, Poorly Sorted | Gravelly Sand | Very Fine Gravelly Coarse Sand |
| SIER2 | Ierissos | IERP-1 | | Bimodal, Moderately Well Sorted | Sandy Gravel | Sandy Very Fine Gravel |
| SOUR4 | Ouranoupoli | | | Bimodal, Poorly Sorted | Slightly Gravelly Sand | Slightly Medium Gravelly Fine Sand |
| SOUR2 | | | | Polymodal, Poorly Sorted | Gravelly Sand | Very Fine Gravelly Coarse Sand |
| SOUR5 | | | | Trimodal, Moderately Sorted | Gravelly Sand | Very Fine Gravelly Very Coarse Sand |

**Figure 2.** Sampling site of sample SSTR1C. The dark brown layers account for heavy mineral concentrations.

2.3 Gamma-ray spectroscopy

The samples after oven-dried at 60°C to constant weight, were 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 first spectrometry system with the High Purity Ge detector was used to measure the majority of the natural radionuclides examined in this study, except $^{238}$U. The second one with the Low Energy planar Ge detector was used so as to determine only the concentration of $^{238}$U, considering the low energy γ-ray of 63 keV emitted by its daughter $^{234}$Th.

The $^{40}$K content was obtained using its 1461 keV γ-ray. The $^{232}$Th content was calculated as the weighted mean value of $^{228}$Ra concentration (measured as $^{228}$Ac, using 911, 968 and 338 keV γ-rays) and $^{228}$Th concentration (measured as decay products in equilibrium, i.e. $^{212}$Pb, using 238 and 300 keV γ-rays, $^{212}$Bi, using 727 keV γ-ray and $^{208}$Tl, using 2614, 583 and 860 keV γ-rays). The determination of $^{226}$Ra content was based on measurement of $^{222}$Rn decay products being in equilibrium. The measurement of $^{226}$Ra from its own γ-ray at 186.25 keV introduces some problems because of the adjacent photo peak of $^{235}$U at 185.75 keV, so that the isotopic ratio between $^{235}$U and $^{238}$U was considered being the natural one, i.e. 0.0072 and

secular equilibrium between $^{238}U$ and $^{226}Ra$ had to be assumed. Accuracy in the measurements of $^{226}Ra$ concentrations by $^{222}Rn$ decay products depended on the integral trapping of radon gas in the sample volume, so a small addition (~2%) of charcoal in powder form (less than 400 μm in size) was mixed with the sample before sealing it hermetically and storing it in a freezer during $^{222}Rn$ in-growth period (Manolopoulou et al. 2002).
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 and self-absorption effects of the emitting gamma photons. A set of high quality certified reference materials (RGU-1, RGTh-1, RGK-1) (I.A.E.A. 1987) was used, with densities similar to the average beach sands measured after pulverization. Cylindrical geometry was used assuming that the radioactivity is homogenously distributed in the measuring samples. The samples were measured up to 200.000 s in order to achieve a Minimum Detectable Activity of 12 Bq $kg^{-1}$ for $^{40}K$, 4 Bq $kg^{-1}$ for $^{232}Th$, 2 Bq $kg^{-1}$ for $^{228}Th$, 2 Bq $kg^{-1}$ for $^{226}Ra$ and 21 Bq $kg^{-1}$ for $^{238}U$, with 33% uncertainty. 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 0.3–2% and the random errors of the radioactivity measurements extend up to 19 %, except in the $^{238}U$ measurement, where the error extends up to 50% for activities measured lower 10 Bq $kg^{-1}$.

## 3. Results and discussion

The location of each sample, the neighboring sample of NE Chalkidiki granitoids which is most probably the main parental rock of the beach sands studied and the textural group of each sample are given in Table 1. The majority of the beach sands studied is moderately or poorly sorted and is classified as gravelly sand. The heavy, heavy magnetic and heavy non-magnetic (% wt.) fractions are given in Table 2.

**Table 2.** Heavy fraction (HF) % wt., heavy magnetic fraction (HMF) % wt., heavy non-magnetic fraction (HNMF) % wt. of the samples studied.

| | **HMF%** | **HNMF%** | **HF%** |
|---|---|---|---|
| SSTR1C | 93.85 | 9.80 | 28.52 |
| SSTR1A | 19.93 | 5.10 | 14.83 |
| SIER2 | 0.03 | 0.02 | 0.01 |
| SOUR4 | 3.06 | 2.52 | 0.54 |
| SOUR2 | 0.90 | 0.72 | 0.17 |
| SOUR5 | 0.01 | 0.00 | 0.01 |

The activity concentrations of $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{228}Th$ and $^{40}K$ (Bq $kg^{-1}$), along with the respective standard errors of the measurements (±σ) are given in Table 3. The activity concentrations of $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{228}Th$ and $^{40}K$ ranged from 9.0-63.0, 11.5-62.9, 10.9-54.5, 11.8-56.6 and 27-828 Bq $kg^{-1}$, respectively. The activity concentration values of the sand samples presented in Table 3, are in well agreement with the natural radioactivity levels reported by the literature concerning data from regular background radiation areas (Freitas and Alencar 2004, Vassas et al. 2006, Harb 2008, Nada et al. 2012).
Uranium and thorium are primarily associated with heavy minerals such as monazite, zircon, and allanite, and secondarily associated with apatite and titanite.
The amount (% wt.) of each of the heavy fractions (total, magnetic and non-magnetic) is correlated with the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ (Fig. 3) presenting moderate positive correlations for $^{238}U$ and scattered but positive correlations for $^{232}Th$. As expected from the fact that the major $^{238}U$ and $^{232}Th$ carrying minerals do not contain K and thus $^{40}K$, $^{40}K$ is negatively correlated with the heavy mineral content. Moreover, $^{238}U$ is better correlated with the heavy magnetic fraction, while $^{232}Th$ is best correlated with the heavy non-magnetic fraction.

The values of the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ of the adjacent rock-types of NE Chalkidiki granitoids (Tab. 4) (Papadopoulos et al. 2013) which are the most probable parental rocks of the beach sands, have been used to normalize the respective values measured for the beach sands. In Figure 4, the normalized values (activity concentration in beach sands/activity concentration in the respective sample of the NE Chalkidiki granitoids) of $^{238}U$, $^{232}Th$ and $^{40}K$ are plotted. Except for sample SOUR4 which is slightly enriched in $^{232}Th$, all the samples are depleted in the natural radionuclides measured. This depletion can be attributed to the enrichment of the beach sands mainly in quartz, which is the mineral being the most abundant in the parental rocks and the most resistant to chemical weathering.

**Table 3.** Activity concentrations of $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{228}Th$ and $^{40}K$ ($Bq \cdot kg^{-1}$), along with the respective standard errors (±σ).

| | **$^{238}$U- series** | | | | **$^{232}$Th- series** | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | **$^{238}$U** | **±σ** | **$^{226}$Ra** | **±σ** | **$^{232}$Th** | **±σ** | **$^{228}$Th** | **±σ** | **$^{40}$K** | **±σ** |
| SSTR1C | 35 | 3 | 58.8 | 0.5 | 12.3 | 0.8 | 12.2 | 0.4 | 27 | 3 |
| SSTR1A | 63 | 4 | 62.9 | 0.7 | 25.8 | 1.3 | 25.6 | 0.6 | 532 | 11 |
| SIER2 | 9 | 5 | 12.0 | 0.4 | 13.0 | 1.2 | 12.1 | 0.5 | 828 | 16 |
| SOUR4 | 31 | 6 | 35.2 | 0.6 | 54.5 | 1.7 | 56.6 | 1.0 | 651 | 12 |
| SOUR2 | 17 | 4 | 18.0 | 0.5 | 27.9 | 1.3 | 27.0 | 0.7 | 685 | 14 |
| SOUR5 | 12 | 6 | 11.5 | 0.3 | 10.9 | 0.9 | 11.8 | 0.4 | 770 | 12 |

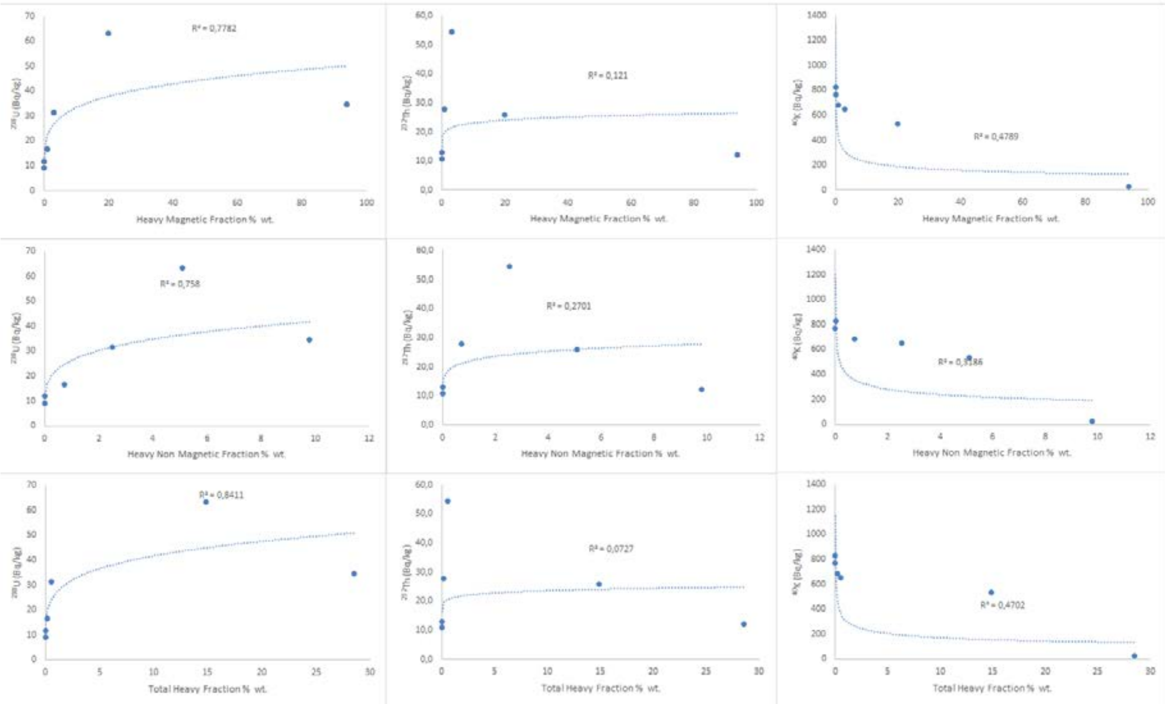


**Figure 3.** Correlations between the activity concentrations of the radionuclides, the (% wt.) total heavy fraction, the heavy magnetic fraction and the heavy non-magnetic fraction

**Table 4.** Activity concentrations of the radionuclides measured in the neighboring granitoid rock-types (Papadopoulos et al. 2013)

| | $^{238}U$ (Bq/kg) | ±σ | $^{232}Th$ (Bq/kg) | ±σ | $^{40}K$ (Bq/kg) | ±σ |
|---|---|---|---|---|---|---|
| **IERP-1 (Ierissos-Ouranoupoli)** | 36 | 5 | 22 | 1 | 748 | 11 |
| **STR-1 (Stratoni)** | 58 | 5 | 80 | 2 | 929 | 13 |

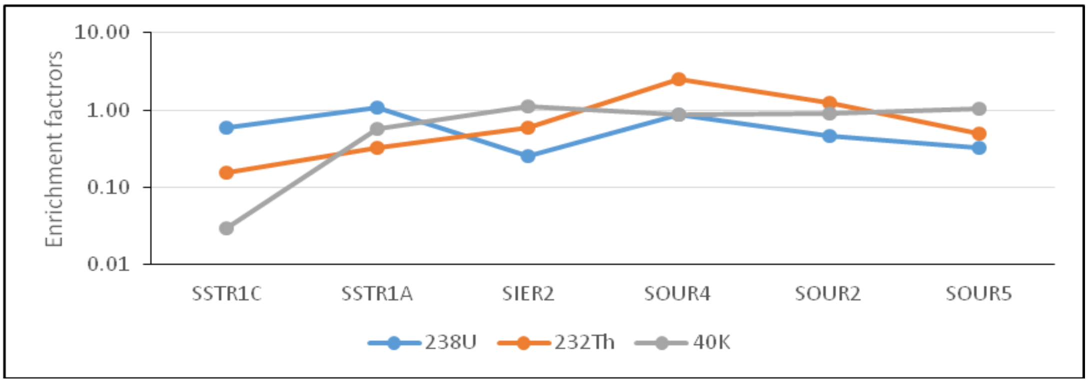


**Figure 4.** Activity concentrations of the natural radionuclides in beach sands normalized to the respective values of the adjacent granitoid rock-types

### 3.1 Exposure dose rates calculations

The radiation received by the population due to beach sands is mainly the result of the time spent on the beach during summer. More specifically, two categories of people were assumed: (a) tourists spending on the beach 8 h per day for 3 weeks and (b) local people working 10 h per day for 10 weeks on the beach.

The absorbed gamma dose rate received implying a uniform distribution of radionuclides, was estimated using the following equation considering the necessary conversion factors (in nGy $h^{-1}$ by Bq $kg^{-1}$) to transform the activity concentrations of $^{40}K$, $^{238}U$ and $^{232}Th$ ($C_K$, $C_U$ and $C_{Th}$ respectively) in absorbed dose rate at 1m above the ground (UNSCEAR 2000, Nada et al. 2012, Velasco et al. 2007, Antovic et al. 2010, Rizzotto et al. 2009, Jabbar et al. 2010, Tsuey-Lin et al. 2011).

$$D_a(nGyh^{-1}) = 0.462 \cdot C_U + 0.604 \cdot C_{Th} + 0.0417 \cdot C_K \qquad (1)$$

The annual effective dose received by the population ($H_{ext}$, mSv/y) was estimated as follows considering the proper conversion coefficient from absorbed dose in air to effective dose (0.7, Sv $Gy^{-1}$) and the outdoor occupancy factor T (h $y^{-1}$):

$$H_{ext}(mSvy^{-1}) = 10^{-6} \cdot D_a \cdot 0.7 \cdot T \qquad (2)$$

The outdoor occupancy factor T ranged from 700 (h $y^{-1}$), for local people working on the beach, down to 168 (h $y^{-1}$) for tourists. The annual effective dose varies between 0.003 and 0.009 mSv $y^{-1}$ for tourists and from 0.012 to 0.037 mSv $y^{-1}$ for local people working on the beach (Tab. 5). The values corresponding to ordinary sand samples are by far lower than the limit of 1 mSv $y^{-1}$.

**Table 5.** Radiation hazard indices calculated due to gamma exposure for the beach sands studied

| **Sample** | **Dair (nGy h-1)** | **Outdoor annual effective dose (mSv y-1)** | |
|---|---|---|---|
| | | **Local people (T = 700 h y-1)** | **Tourists (T = 168 h y-1)** |
| SSTR1C | 24.56 | 0.012 | 0.003 |
| SSTR1A | 67.01 | 0.033 | 0.008 |
| SIER2 | 46.58 | 0.023 | 0.005 |
| SOUR4 | 74.62 | 0.037 | 0.009 |
| SOUR2 | 53.10 | 0.026 | 0.006 |
| SOUR5 | 44.18 | 0.022 | 0.005 |

## 4. Conclusions

Beach sands adjacent to the Stratoni, Ierissos and Ouranopolis granites demonstrate heavy fractions (total, magnetic and non-magnetic; in wt. %) presenting moderate positive correlations for $^{238}$U and scattered but positive correlations for $^{232}$Th. Moreover, $^{238}$U is better correlated with the heavy magnetic fraction, while $^{232}$Th is best correlated with the heavy non-magnetic fraction.

All the samples are depleted in the natural radionuclides measured. This depletion can be attributed to the enrichment of the beach sands mainly in quartz, which is the mineral being the most abundant in the parental rocks and the most resistant to chemical weathering.

The annual effective dose varies between 0.003 and 0.009 mSv $y^{-1}$ for tourists and from 0.012 to 0.037 mSv $y^{-1}$ for local people working on the beach. The values corresponding to ordinary sand samples are orders of magnitude lower than the limit of 1 mSv $y^{-1}$ even for sample SSTR1C which is heavy mineral enriched. Therefore, the maximum value of annual effective dose is by far lower than the permitted value of 1 mSv $y^{-1}$ in the case of all samples.

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