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Natural radioactivity distribution and gamma radiation exposure of beach sands close to Maronia and Samothraki plutons, NE 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 of Maronia and Samothraki being adjacent to the local granitoids. These range from 14-75, 17.5-114.8, 23.7-71.9, 24.5-71.6 and 262-1319 Bq/kg, respectively. The (wt.%) of heavy magnetic (HM) (amphibole, epidote, clinopyroxene, chlorite, magnetite and ilmenite) fraction, the heavy non-magnetic (HNM) (zircon and titanite) 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 0.004 and 0.013 mSv y^{-1} for tourists and from 0.016 to 0.056 mSv y^{-1} for local people working on the beach.

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Key words: natural radioactivity, beach sands, granitic rocks, heavy magnetic and non-magnetic fractions, annual effective dose equivalent, NE Greece.

INTRODUCTION

Beach sands are mainly composed 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, 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 for health care 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 such as India and Brazil or countries with extended sand landscapes. Exposure dose rates of the public have also been assessed indicating that these dose rates vary depending upon the concentration of the natural radionuclides present in sands and bed-rocks, which in turn depends 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, 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 are 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).

This study is focused on the beach sands that are placed adjacent to the rock-types of the granitoids of Samothraki Island and Maronia, which are their most probable parental rock. All the 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. Relations of several mineral fractions separated from the whole sample (total heavy, heavy magnetic and heavy non-magnetic wt.%), and the content of natural radio-nuclides are also assessed. Evaluation of the enrichment or depletion of the natural radionuclides in beach sands relative to the parental granitic 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 are also presented.

GEOLOGICAL SETTING

Both Maronia and Samothraki plutons intrude the Circum-Rhodope belt.

Maronia pluton

Three main rock groups have been distinguished for the Maronia pluton: a) the basic; b) the intermediate and; c) the acid group. The basic group is composed of gabbro, a dark coloured medium grained rock having clinopyroxene, red-brown biotite, plagioclase, and Fe-Ti oxides. The rocks of the intermediate group include monzonite, quartz monzonite, monzogabbro, quartz monzogabbro and mafic micogranular enclaves. The acid group consists of granite, porphyritic micro-granite and aplitic dykes. The rocks of the basic group show a high-K, calc-alkaline affinity while those of the intermediate group are characterized as shoshonites. The rocks of the acid group are considered as ultra-potassic (Papadopoulou et al., 2004). The age of the Maronia pluton, based on Rb-Sr whole rock-biotite isochrones, has been determined as Oligocene (29 Ma) and is considered to be the youngest of the Tertiary plutons that intruded the Circum Rhodope belt (Del Moro et al., 1988).

Samothraki pluton

The Samothraki pluton is an intrusion composed of hb-bi quartz monzonite, bi-hb quartz monzonite, bi-hb granite and bi granite. All the above mentioned rock-types have porphyritic texture. The mineral constituents of the rock types are quartz, feldspars, biotite, hornblende, magnetite, ilmenite, apatite, zircon, titanite, allanite and rutile. It is high-K alkaline, metaluminous to slightly peraluminous intrusion and is considered to have been emplaced in an extensional environment. The Rb-Sr biotite-whole rock cooling age of the pluton is 18.5 Ma (Christofides et al., 2000).

COLLECTION AND PRE-TREATMENT OF THE SAMPLES

Representative sediment samples obtained from 6 random points (separated each other by around 500 m) were collected from beaches close to the granitoid rocks of Maronia and Samothtraki (Fig. 1, Table 1) using sampling intervals relative to the length of each beach.

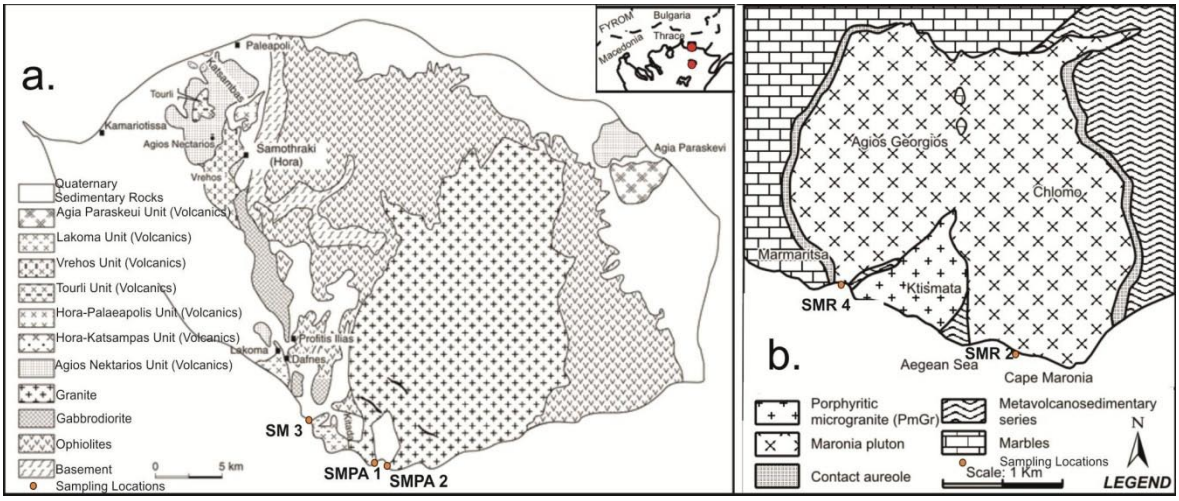

Fig. 1. Geological sketch of: a. Samothraki (Vlahou, 2003); and b. Maronia (Papadopoulou et al., 2004) plutons presenting the sampling sites.

Two samples (SM3 and SMR4) were collected from a specific horizon of green color and thickness of 1-4 cm, enriched in heavy minerals, especially epidote (Table 1, Fig. 2).

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

In the laboratory, the samples were cleaned with warm water and dried. Coarse wastes (e.g. sea shells) 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, epidote, clinopyroxene, chlorite, magnetite and ilmenite while the heavy non-magnetic fraction (>0.8 amp at same settings) contains zircon and titanite. 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.

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 consists of a 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 consists 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 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

Table 1.
Location, sample and textural characteristics of the samples studied

Sample	Location (beach name)	Comments	SAMPLE TYPE	TEXTURAL GROUP	SEDIMENT NAME
SMR2	Maronia	Heavy minerals horizon	Trimodal, Very Poorly Sorted	Gravelly Mud	Very Fine Gravelly Fine Silt
SMR4			Trimodal, Poorly Sorted	Slightly Gravelly Sand	Slightly Very Fine Gravelly Coarse Sand
SM3	Samothraki	Heavy Minerals horizon	Bimodal, Poorly Sorted	Gravelly Sand	Very Fine Gravelly Fine Sand
SMPA2			Bimodal, Moderately Well Sorted	Very Fine Gravel	Gravel
SMPA1			Polymodal, Poorly Sorted	Gravelly Sand	Very Fine Gravelly Coarse Sand

Fig. 2. Sampling sites of samples SM3 from Samothraki and SMR4 from Maronia.

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

	HMF	HNMF	HF
SMR2	8.14	0.11	8.03
SMR4	62.63	0.86	61.77
SM3	12.93	0.53	12.40
SMPA2	0.06	0.01	0.05
SMPA1	0.01	0.00	0.01

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 measured 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 range 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}.

X-RAY DIFFRACTION SPECTROSCOPY

Three samples were submitted for mineral identification and semi-quantitative analysis at ACTLABS, Ontario, Canada. These were heavy magnetic and heavy non-magnetic fractions of several beach sand samples (Table 3).

A portion of each pulverized sample was loaded into a standard holder. The quantities of the crystalline mineral phases were determined using the Rietveld method. It is based on the calculation of the full diffraction pattern from crystal structure information. The X-ray diffraction analysis was performed on a Panalytical X'Pert Pro diffractometer, equipped with a Cu X-ray source and an X'Celerator detector, operating at the following conditions: 40 kV and 40 mA; range 5–70 deg 2θ; step size 0.017 deg 2θ; time per step 50.165 sec; fixed divergence slit, angle 0.5^{o}; sample rotation 1 rev/sec.

RESULTS AND DISCUSSION

The location and the textural group of each sample are given in Table 1. The majority of the beach sands studied is poorly sorted and is classified as gravelly sand. The heavy, heavy magnetic and heavy non-magnetic (wt.%) fractions are given in Table 2.

The activity concentrations of ^{238}U, ^{226}Ra, ^{228}Ra,

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
SMR2	38	4	43.7	0.4	44.9	1.2	42.3	0.9	791	13
SMR4	75	5	114.8	0.9	71.9	2.0	71.6	1.2	262	7
SM3	14	5	17.5	0.4	23.7	1.1	24.5	0.6	298	8
SMPA2	57	6	49.6	0.5	58.7	1.5	58.0	0.9	1245	18
SMPA1	52	7	46.8	0.7	54.9	2.0	56.1	1.1	1319	22

Table 4.
Semi-quantitative mineralogical composition of the heavy magnetic fraction (HM)

	SMR2HM	SMR4HM	SM3HM
Amphibole	43	18	50
Epidote	n.d.	n.d.	38
Clinopyroxene	50	n.d.	4
Chlorite	n.d.	n.d.	8
Magnetite	7	72	n.d.
Ilmenite	n.d.	10	n.d.

^{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-series, ^{232}Th-series and ^{40}K range from 14-75, 17.5-114.8, 23.7-71.9, 24.5-71.6 and 262-1319 Bq kg^{-1}, respectively. The values of the sand samples presented in Table 1 are in good agreement with the natural radioactivity levels reported in the literature, concerning data from regular background radiation areas (Freitas, 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 (Papadopoulos et al., 2014). However, as it is indicated in Table 2, the heavy non-magnetic fraction that contains monazite and zircon is present in very small amounts in the investigated samples. What is more, allanite is not present in none of the heavy magnetic fraction of the samples studied (Table 4).

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 scattered positive correlations for ^{238}U and ^{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. Similar trends but certainly higher correlation coefficients have been reported by Papadopoulos et al. (2014) for heavy minerals enriched beach sands of Sithonia. However, it has to be noted that the statistics of the present work are based on only 5 samples.

The average values of the activity concentrations of ^{238}U, ^{232}Th and ^{40}K of the adjacent rock-types of Maronia and Samothraki plutons (Table 5) (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

Table 5.
Average 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
Maronia	111.9	5.9	137.6	3.3	959	12
Samothraki	144.5	6.5	109.5	2.5	896	3

Table 6
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 ($T = 168\ h\ y^{-1}$)
SMR2	77.44	0.038	0.009
SMR4	89.10	0.044	0.010
SM3	33.40	0.016	0.004
SMPA2	113.65	0.056	0.013
SMPA1	112.37	0.055	0.013

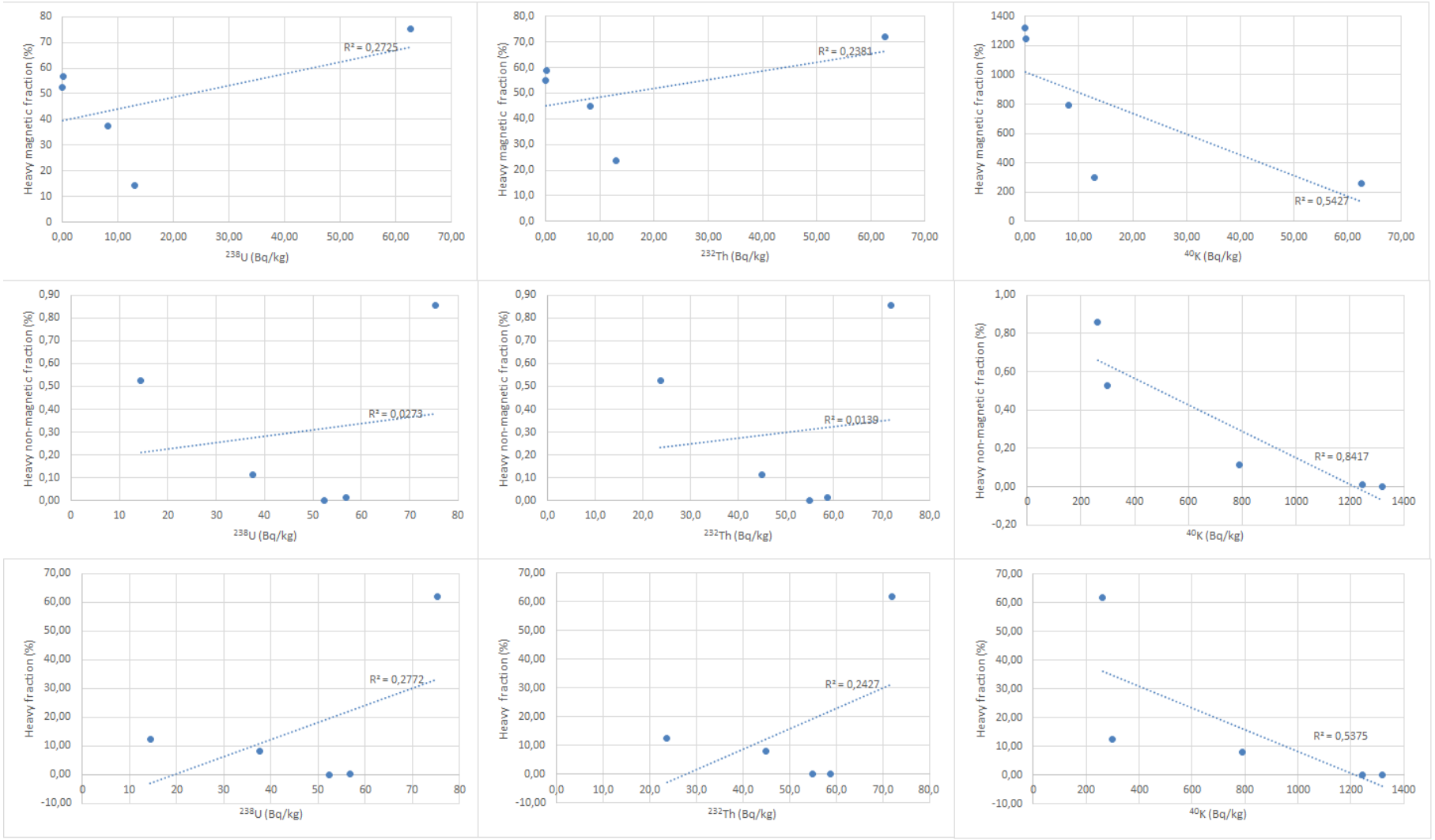

Fig. 3. Correlations between the activity concentrations of the radionuclides, the (wt. %) total heavy fraction, the heavy magnetic fraction and the heavy non-magnetic fraction.

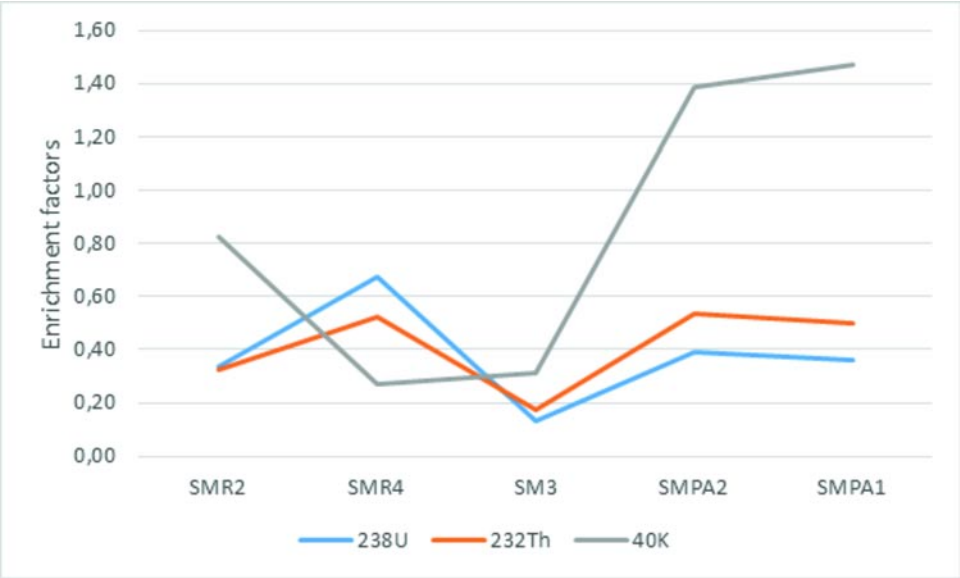

Fig. 4. Activity concentrations of the natural radionuclides in beach sands normalized to the respective values of the adjacent granitoid rock-types.

the average of the granitoids) of ^{238}U, ^{232}Th and ^{40}K are plotted. Except for samples SMPA1 and SMPA2 from Samothraki which are slightly enriched in ^{40}K, 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. Moreover, the depletion can be due to the fact that the mineral constituents of the heavy magnetic fractions (amphibole, clinopyroxene, chlorite, magnetite, ilmenite and epidote) do not contain U and Th in considerable amounts in their crystal lattice. Moreover, the wt.% heavy non-magnetic fractions that contain mainly zircon are very limited.

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 the 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).

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

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}):

$$H_{ext}\,(mSvy^{-1}) = 10^{-6}\,.\,D_a\,.\,0.7\,.\,T \quad (2)$$

The outdoor occupancy factor T ranges 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 0.004 and 0.013 mSv y^{-1} for tourists and from 0.016 to 0.056 mSv y^{-1} for local people working on the beach (Table 5). The values corresponding to ordinary sand samples are by far lower than the limit of 1 mSv y^{-1}.

CONCLUSIONS

The amount (wt.%) of each of the heavy fractions (total, magnetic and non-magnetic) presents weak positive correlations for ^{238}U and ^{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. Moreover, the heavy minerals contained in the investigated samples do not contain considerable amounts of U and Th and thus any minerals that include them in their crystal lattice.

The annual effective dose varies between 0.004 and 0.013 mSv y^{-1} for tourists and from 0.016 to 0.056

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 SM3 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 and no risk is involved for human population.

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