NATURAL RADIOACTIVITY DISTRIBUTION AND GAMMA RADIATION EXPOSURE OF BEACH SANDS CLOSE TO THE GRANITOIDS OF NORTHERN GREECE

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ABSTRACT

Compared to other natural stones, granitoid rocks are well known for their elevated natural radioactivity levels. This study aims to evaluate the activity concentrations of ^{238}U and ^{226}Ra from ^{238}U-series, ^{232}Th and ^{228}Th from ^{232}Th-series as well as ^{40}K along the beaches of northern Greece, being adjacent to local granitoids. More specifically, these beaches are located at Sithonia Peninsula, NE Chalkidiki (Ierissos, Stratoni, Ouranoupoli), Kavala and NE Greece (Maronia and the island of Samothraki). Some of the investigated samples were considerably enriched in heavy minerals. In particular, these were allanite, epidote, amphibole, mica, clinopyroxene, magnetite, ilmenite, pyrite, monazite, zircon, titanite, arsenopyrite and apatite. Among them, monazite and allanite contain more than a few percent of U and especially Th in their crystal lattice. Considering that 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 equivalent dose was estimated. The values corresponding to ordinary (non-heavy minerals-enriched) sand samples are orders of magnitude lower than the limit of 1 mSv y^{-1} while in the case of heavy minerals-rich sands and more specifically a sample from Kavala which contains 81.05% wt. heavy minerals, the annual equivalent dose is estimated up to 1.48 mSv y^{-1}.

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

1. Introduction

Beach sands are mineral deposits formed through the combination of weathering, fragmentation and degradation. They are composed mainly of quartz, feldspar and other minerals resistant to wave abrasion.

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, 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 (Alam *et al*., 1999; Shetty *et al*. 2011; Nada *et al*., 2012)

The coastline of Greece is one of the largest worldwide. Despite of the fact that most of Greek beaches are highly touristic, there are few 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 northern Greece [Sithonia Peninsula, NE Chalkidiki (Ierissos, Stratoni and Ouranoupoli), Kavala and NE Greece (Maronia and the island of Samothraki)], which are their most probable parental rocks. The goal of this work is to assess the activity concentrations of the natural radionuclides in relation with the heavy minerals fractions. Moreover, data about the annual equivalent dose received by the population due to their exposure on gamma radiation by the beach sands are presented.

2. Materials and methods

2.1. Collection and pre-treatment of the samples

Representative sediment samples obtained from 58 random points from Sithonia (Papadopulos *et al*. 2014), 37 from Kavala, 6 from NE Chalkidiki (Ierissos, Stratoni and Ouranoupoli) and 5 from Maronia and Samothraki (separated each other by around 500m). All of them were close to the local granitoid rocks of the respective areas, using sampling intervals relative to the length of each beach. Several samples from Sithonia, Kavala, Stratoni, Maronia and Samothraki were collected from specific horizons and thickness of 1-4 cm, enriched in heavy minerals.

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. 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) was used to take the heavy mineral fractions. The mineral content of the heavy fractions was allanite, epidote, amphibole, mica, clinopyroxene, magnetite, ilmenite, pyrite, arsenopyrite, monazite, zircon, titanite and apatite. All the above mentioned minerals have been identified under the binocular microscope and SEM-EDS. 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.

2.2. Gamma-ray spectroscopy

The samples after oven-dried at 60°C to constant weight, were measured using two high-resolution γ-ray spectrometry systems. The first one consisted of an HPGe coaxial detector with 42% efficiency and 2.0keV 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 0.7keV resolution at 122keV 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 to determine only the concentration of ^{238}U, considering the low energy γ-ray of 63keV 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 ranges of the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Th and ^{40}K for the samples from Sithonia were 6-673.0, 5-767, 5-1750, 6-1760 and 185-875 Bq kg^{-1}, respectively. For the samples from Kavala they were 14-940, 16-1710, 26-4547, 27-4488 and 194-1307 Bq kg^{-1}. For the samples from NE-Chalkidiki they were 9.0-63.0, 11.5-62.9, 10.9-54.5, 11.8-56.6 and 27-828 Bq kg^{-1} respectively. Finally for the samples from NE Greece they were 14-75, 17.5-114.8, 23.7-71.9, 24.5-71.6 and 262-1319 Bq kg^{-1}. The values of the sand samples that are not enriched in heavy minerals, are in well agreement with the natural radioactivity levels reported by the literature concerning data from regular background radiation areas.

Uranium and thorium are primarily associated with heavy minerals such as monazite and allanite (Papadopoulos *et al*., 2014). These are present in the heavy minerals-enriched sands of Sithonia and Kavala. Certain samples from Maronia, Stratoni and Samothraki are enriched in heavy minerals, but the mineral constituents of their heavy fractions involve amphibole, clinopyroxene, chlorite, magnetite, ilmenite, epidote, pyrite and arsenopyrite that do not contain U and Th in their crystal lattices.

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; Antovic *et al*., 2010; Jabbar *et al*., 2010; Tsuey-Lin *et al*., 2011; Nada *et al*., 2012).

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

The annual equivalent dose for whole body irradiation (w_{Tissue}=1) received by the population (E_{ext}, mSv/y) was estimated as follows considering the proper conversion factor from absorbed dose (Da, Gy) in air to dose equivalent (Hext, Sv) and the outdoor occupancy factor T (h y^{-1}):

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

Assuming that the mean energy of gamma lines emitted by ^{238}U- and ^{232}Th-series as well as the gamma line of ^{40}K ranged from 0.66 up to 1.46 MeV then a conversion factor of 0.7 Sv.Gy^{-1} can be taken into account as a representative one (Keith *et al*., 1993). 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.

According to Papadopoulos *et al*. (2014), the annual equivalent dose for the beach sands from Sithonia varies between 0.003 and 0.165 mSv y^{-1} for tourists and from 0.013 to 0.688 mSv y^{-1} for local people working on the beach. The respective values for the samples from Kavala beach sands vary between 0.01 and 0.35 mSv y^{-1} for tourists and from 0.03 to 1.48 mSv y^{-1} for local people working on the beach. The range for beach sands from NE Chalkidiki (Ierissos, Stratoni, Ouranoupoli) is 0.003-0.009 mSv y^{-1} for tourists and 0.012-0.037 mSv y^{-1} for local people working on the beach. As for beach sand samples from NE Greece (Maronia, Samothraki), the range is 0.004-0.013 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 (non-heavy minerals-enriched) are orders of magnitude lower than the limit of 1 mSv y^{-1}. However, in the case of heavy minerals-rich sands and more specifically a sample from Kavala which contains 81.05% wt. heavy minerals and ≈5% wt. of allanite, the annual equivalent dose is estimated up to 1.48 mSv y^{-1}.

4. Conclusions

Generally, the concentrations of the radionuclides measured are elevated in the heavy minerals-enriched samples. In particular, this is true only for samples that contain considerable amounts of allanite, monazite and zircon in their heavy fractions. So, natural radioactivity of beach sands is related with the presence of certain minerals of the heavy fractions.

The values corresponding to ordinary sand samples (non-heavy minerals-enriched) are orders of magnitude lower than the limit of 1 mSv y^{-1} while in the case of heavy minerals-enriched sands and more specifically for a sample from Kavala (containing 81.05% wt. heavy minerals and ≈5% wt. allanite), the annual equivalent dose for local people is estimated up to 1.48 mSv y^{-1}. Therefore, at least for tourists it seems that the maximum value of annual equivalent dose is by far lower than the permitted value of 1 mSv y^{-1} in the case of most of the samples. However, in terms of as low as reasonably achieved exposure to natural radioactivity, the beaches of Kavala and Sithonia where heavy minerals enriched horizons are present should preferably be avoided.

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