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Radionuclides in rocks of southern part of Mtskheta-Mtianeti region (Georgia)

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ABSTRACT

This study investigates distribution of natural and technogenic radionuclides in rock samples in the southern part of the Mtskheta-Mtianeti region, located in the so-called Transcaucasian intermountain area (II) and in the Lesser Caucasus fold system (III). Several geotectonic units are identified here: the eastern immersion zone or Georgian Block (II_3), the molassic subzone of Kartli (II_3^1), the central (axial) subzone (III_1^3), and the southern subzone (III_1^4) of the Adjara-Trialeti folded zone (III_1). The radioactivity of rocks in this region has not been investigated. Twenty samples of rocks of various types, including sedimentary and metamorphic, were investigated with the gamma-spectroscopic method. Up to 21 naturally occurring radionuclides and one technogenic radionuclide were identified in samples. Average activity concentration of Th-232 family radionuclides varied from 4.5 to 67.0 Bq/kg, U-238 family: from 4.5 to 93.5 Bq/kg, and U-235 family: from 0.22 to 4.3 Bq/kg. The highest activity concentration was observed for K-40 (maximal value of 746 Bq/kg). Activity of technogenic radionuclide Cs-137 varied from 0.12 to 6.3 Bq/kg. There are some marked features in radionuclide distribution depending, in particular, on rock type, tectonic zone, and age. Some radionuclide activity ratios have been considered: U-238/U-235, U-238/Th-232, Ra-226/U-238, and Pb-210/Ra-226. These ratios allowed estimation of the system's condition (closed or opened) and information about the character of relevant geochemical processes. Comparison was carried out with existing data in the literature.

1. Introduction

Natural radioactivity in the environment is caused by radionuclides of three families: Th-232, U-238, and U-235, as well as K-40, which are so-called alpha-, beta-, or gamma-emitters. Long-lived radionuclide Cs-137 (as well as Sr-90) is the most widespread among technogenic radionuclides. Research on the radioactivity of rocks is a subject of numerous studies.

Rafique et al. (2014) studied samples of sedimentary (limestone, sandstone, and red shale) and metamorphic (black shale, slate, graphite, marble, quartz mica, calcareous schist, and quartz) rocks from the state of Azad Kashmir (Pakistan). In the samples of sedimentary rocks, activity concentration of Ra-226 varied from 18.1 to 45.2 Bq/kg (average value of 31.4 Bq/kg), Th-232 – from 28.7 to 68.9 Bq/kg (average value of 48.5 Bq/kg), and K-40 – from 224 to 610 Bq/kg (average value of 381 Bq/kg). Authors stated that the activity concentration of Cs-137 for all the samples remained below the lower limit of detection of the system. Estimation of radiological hazards was carried out; in particular, values of radium equivalent activity for all investigated samples varied from 20.6 to 294 Bq/kg (average activity value was 126 Bq/kg), which is sufficiently below the recommended value of 370 Bq/kg (UNSCEAR, 1988; ICRP, 2007).

Frančišković-Bilinski et al. (2004) studied samples of tuff deposits from four separate occurrences in the Dinaric Karst of Croatia. The activity concentration of Th-232 varied from 1.0 to 12.9 Bq/kg with average value of 4.5 Bq/kg; average value of U-238 concentration was sufficiently greater at 7.5 Bq/kg (from 1.3 to 14.6 Bq/kg); Ra-226 had an average value of 7.3 Bq/kg – from 2.9 to 13.0 Bq/kg. In the investigated samples, the technogenic radionuclide Cs-137 was also detected, whose activity varied within the interval 3.9–77.8 Bq/kg with an average value of 24.6 Bq/kg. The authors describe it as Cs-137 impurity originating from rainwater. Outdated tuff is not a closed system and sampling was performed after an accident in 1986, followed by heavy rain in this region. Activity ratios were also calculated for U-238/

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Th-232, which varied from 0.81 to 5.30 with average value of 2.41, and for Ra-226/U-238 – from 0.59 to 2.22 with average value of 1.10.

Various rocks used as building materials in Romania were studied in the work (Pantelica et al., 2001). Among them was tuff. The activity concentrations of Th-232, Ra-226, K-40, and Cs-137 in the tuff were 9.8, 10.5, 180, and 9.6 Bq/kg, respectively.

Turhan (2010) studied some tens of limestone samples collected from all over Turkey. The activity concentration of Ra-226 varied from 0.7 to 55.1 Bq/kg with an average value of 19 Bq/kg, Th-232 – from 1.2 to 20.9 Bq/kg with an average value of 4.3 Bq/kg, and K-40 – from 10.1 to 258.4 Bq/kg with an average value of 55 Bq/kg. Calculated values of equivalent activity varied within the interval 3.2–81.6 Bq/kg with an average value of 29.4 Bq/kg.

Research on various environmental objects' radioactivity in Georgia has been carried out in the past, stimulated by the meltdown at the Chernobyl atomic power station in 1986. The elevated concentrations of various technogenic radionuclides (up to several thousands of Bq/kg) were observed especially in soil of the western Georgian coastal region (Nadareishvili et al., 1991).

The present work analyses the natural and technogenic radioactivity of rocks in the territory of Georgia, mainly in the flat part of the Mtskheta-Mtianeti region. Some results of soil radioactivity researches in this region are given in the work (Kekelidze et al., 2017).

2. Materials and methods

2.1. Study area

2.1.1. Area geology

The southern part of the Mtskheta-Mtianeti region is located to the south of the Greater Caucasus fold system (I), in the so-called Transcaucasian intermountain area (II) (up to the territory of the city of Tbilisi) and in the Lesser Caucasus fold system (III) (Gamkrelidze, 2013). In this area, the mountain system of the Great Caucasus main ridge (Kakhetian ridge) is contiguous with the Lesser Caucasus mountain system (Trialeti ridge). Using tectonic characteristics, it is possible to discriminate two areas in this region:

- The eastern immersion zone or Georgian Block (II_3), a region identified as part of the molassic subzone of Kartli (II_3^1). It contains the Mukhrani-Tirifoni and Bazaleti blocks and is a low-mountain (about 500 m above sea level) area near the territory of the city of Mtskheta.
- The Adjara-Trialeti folded zone (III_1) is a mountain area (600–1000 m above sea level) to the southwest of Mtskheta. Two subzones are contiguous in this region (near Dzegvi and Kvemo Nichbisi settlements):
  - o the central (axial) subzone (III_1^3);
  - o and the southern subzone (III_1^4), which has not been investigated at this point.

Metamorphic volcanic and sedimentary rocks (limestone, sandstone, argillite, clay, and slate) are predominant in these areas.

2.1.2. Locations and samples

Twenty rock samples (Table 1) were collected in the investigated area at seventeen locations (located near Bodorna (Bd) and Zedazeni (Zd) settlements; Jvari (Jv) monastery; and Mukhatgverdi (Mg), Karsani (Ks), Armazi (Am), Dzegvi (Dz), Saskhori (Ss), Khekordzi (Kk), and Nichbisi (Nb) settlements), in particular:

- in subzone II_3^1: fourteen samples (22, 23, 68, 66, 90, 70, 71, 72, 89, 86, 87, 85, 84, 79) were collected from twelve locations;
- in subzone III_1^3: six samples (78, 76, 77, 74, 80, 82) were collected from six locations.

Types of collected samples are the following:

- o sedimentary, eighteen samples including:
  - carbonate (limestone): three samples (23, 74, 80);
  - clay: two samples (72, 87);
  - sandstone: thirteen samples (carbonate sandstone: 22, 85; arkosic sandstone: 66, 68, 79, 84; tuff sandstone: 70, 71, 78, 82, 86, 89, 90);
- o metamorphic volcanic: two samples (tuff: 76, 77).

Fig. 1 shows layout of locations. Fig. 2 shows location Ss-2 located on the boundary of two subzones, II_3^1 and III_1^3, where two samples, 79 and 78, were collected.

2.2. Sampling and analysis

2.2.1. Sampling

Samples were collected from the outcrops and placed directly into plastic containers (volume up to 2.0 L). After drying in laboratory conditions, samples were broken into pieces of up to 40 mm, and then were crushed using special crusher (Retsch jaw crusher) to sizes on the order of 1 mm. Then samples were dried at 105–110 °C to constant weight, and their bulk density and weight were determined. These values were used in the description of sample geometry. The samples were sealed in Marinelli beakers (polyvinyl chloride adhesive tape was used for hermetic sealing) and stored for more than four weeks to achieve secular equilibrium between Ra-226 and Rn-222.

2.2.2. Measurement of gamma radiation activity

Measurements were carried out using a Canberra GC2020 gamma spectrometer with a semi-conductor germanium detector with a relative efficiency 24%. Gamma spectra acquisition time was 72 h. For analysis, Genie-2000 S500 software was used with additional modules, in particular, S506 – Interactive Fit Program. This program, for example, carried out “decomposition” of interference peaks for all spectra in the area of 186 keV (the program identified one peak in this area that grew out of the interference of two closely spaced peaks, U-235 [185.715 keV] and Ra-226 [186.211 keV]). S506C mathematically processes a spectral curve and, in this area, two Gaussian peaks with energies corresponding U-235 and Ra-226 were created. During the program's identification of peaks and calculation of activity concentration, a tolerance value was established in such manner that the low-energy peak was compared only with U-235, and high-energy only with Ra-226. The results showed, in particular, a determination error of activity concentration of Ra-226 was mainly within 9–24%. Its activity was compared with the activity of its daughters, Pb-214 and Bi-214, which determination error put in the limits of 3 to 9%. The reported values of activity of Ra-226, Pb-214, and Bi-214 were not different enough from each other. Thus, it is possible to consider, that a determination error of Ra-226 concentration calculated in such a way is satisfactory enough (on occasion, in low-activity samples, activity of Ra-226 was adjusted by value of the average activity of Pb-214 and Bi-214, and the received value was considered at a definitive estimation of activity value of U-235). This method also was used for determination of U-235 activity concentration at 185.715 keV. Reported values of U-235 activity (which determination error on the whole varied from 8 to 16%, and sometimes for low-activity samples – from 18 to 31%) were compared to values of U-238 activity (which was determined by the line at 63.3 keV of Th-234 with an error of 6.3–14%). The value of their activities ratio, U-238/U-235, which is considered constant (21.7) for natural objects, was used as a criterion (UNSCEAR, 1993). When deviations from this value was big (> 10%) the repeated analysis of a line 186 keV (and also line 63.3 keV) by means of program S506 was carried out. Concentration of Pb-210 was determined by the line 46.54 keV (with determination error from 8.3 to 20.6%). To determine Th-232 activity, average values for Ac-228, Ra-224, Pb-212, Bi-212, and Tl-208