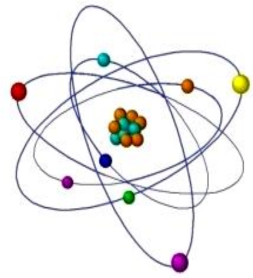
EVALUATION OF RADIOACTIVITY OF THE ROCKS IN THE FOOTHILLS OF THE MAIN CAUCASIAN RANGE IN THE TERRITORY OF KAKHETI REGION (East Georgia-Country)



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ABSTRACT: *This study investigates distribution of natural and technogenic radionuclides in rock samples in the eastern part of Kakheti region. The radioactivity of rocks in this region has not been investigated. 20 samples of rocks of various types – igneous, sedimentary and metamorphic were investigated with the gamma-spectroscopic method applied. Up to 21 naturally occurring radionuclides and 1 technogenic radionuclide have been identified in samples. Average activity concentration of Th-232 family radionuclides varied from 0.61 to 98.6 Bq/kg, U-238 family - from 9.1 to 77.0 Bq/kg, U-235 family - from 0.10 to 3.5 Bq/kg. The highest activity concentration was observed for K-40 (maximal value of 1475 Bq/kg). Activity of technogenic radionuclide Cs-137 varied from 0.10 to 1.2 Bq/kg. There are some marked features in radionuclides distribution depending, in particular, on rock type, tectonic zone. Some radionuclide activity ratios have been considered, in particular, U-238/U-235, U-238/Th-232, Ra-226/U-238 and Pb-210/Ra-226. These ratios allow to estimate condition of system (closed or opened) and to receive a certain notion about character of relevant geochemical processes. Comparison was carried out with existing data in the literature.*

Key words: radionuclides, rocks, radioactivity, activity ratios, East Georgia

INTRODUCTION

The ionizing radiation from the rocks is one of the main natural sources of the radioactivity. Geological bodies with the raised content of naturally occurring radioactive materials (NORM) concern a category of natural geological objects which define ecological conditions of the territory. As is known, natural radioactivity in the environment is caused by radionuclides of three families - Th-232, U-238 and U-235, and also K-40 which are so-called alpha-, beta- or gamma-emitters. Long-lived radionuclide Cs-137 (as well as Sr-90) is most widespread among technogenic radionuclides. Researches of radioactivity of rocks are a subject of numerous studies.

Previous work [1] has focused on rock samples selected in İkizdere and Kaptanpaşa Valley, Turkey. As a result of this work it was determined that, the activity concentrations of rock samples range from 26.12±3.24 to 44.39±4.41 Bq/kg for Th-232, 13.80±1.91 to 29.25±5.04 Bq/kg for U-238 and from 307.54±36.10 to 539.15±22.54 Bq/kg for K-40. The overall mean activity levels for Th-232, U-238 and K-40 are 35.06±3.85, 19.71±2.15 and 386.16±27.38 Bq/kg, respectively. In a different study [2] samples of igneous rocks (dolerite, granite, and granitic gneisses), sedimentary rocks (shale, limestone, sandstone) and metamorphic rocks (black shale, slate, graphite, marble, quartz mica, calcareous schist, and quartz) selected in the state of Azad Kashmir, Pakistan were studied. As a result of this work it was determined that, in samples of metamorphic rocks in particular activity concentration of Ra-226 varied from 5.4 to 100 Bq/kg; concentration of Th-232 varied from ≤1.8 to 111 Bq/kg; and K-40 varied from 9.6 to 1055 Bq/kg. An estimation of radiation hazard was made; in particular the value of the radium equivalent activity of all tested samples varied in the range from 20.6 to 294 Bq/kg, which is sufficiently lower than the recommended limit of 370 Bq/kg [3, 4].

In the work [5] there were studied samples of tuff deposits from four separate occurrences in the Dinaric Karst of Croatia. It was founded that activity concentration of Th-232 varied in the diapason 1.0-12.9 Bq/kg with average value of 4.5 Bq/kg; average value of U-238 concentration was sufficiently greater and made 7.5 Bq/kg (diapason 1.3-14.6 Bq/kg); comparable with U-238 results was obtained for Ra-226 – average value of 7.3 Bq/kg, diapason – from 2.9 to 13.0 Bq/kg. In investigated samples it was also detected technogenic radionuclide Cs-137 which activity varied within the interval 3.9-77.8 Bq/kg with average value of 24.6 Bq/kg; authors explain it as 137Cs impurity originating from rainwater; outdated tuff is not a closed system and sampling was performed after the accident in 1986, followed by heavy rain in this region. Also activities ratios was calculated which values for U-238/Th-232 varied in the diapason 0.81-5.30 with average value of 2.41, and for Ra-226/U-238 – in the diapason 0.59-2.22 with average value of 1.10.

A study in the Modane-Aussois region (Western Alps, France) [6] studied radionuclides content of various characteristic rocks, in particular, sedimentary (carbonaceous breccia, limestone dolomite, dolomite) and metamorphic (calcschist, marble, quartzite) rocks. The activity concentrations of K-40 varied from 18 Bq/kg (limestone dolomite) to 572 Bq/kg (quartzite). The activity concentration of Th-232 varied from < 1 Bq/kg (limestone dolomite) to 18 Bq/kg (calcschist). The highest concentration of U-238 was 29 Bq/kg (dolomite) and the lowest was 9.5 Bq/kg (quartzite).

Researches of various environmental objects radioactivity in Georgia were carried out in the past and, basically, were stimulated by the breakdown which took place on the Chernobyl atomic power station in 1986. The raised concentration of various technogenic radionuclides were observed (up to several thousands of Bq/kg), especially in soil of the West Georgia coastal part [7]. Some results for last period are given in the works [8, 9].

This study represents results of the investigations of the radioactivity of rocks samples selected in the foothills of the Main Caucasian Range in the territory of the region of Kakheti (East Georgia).

MATERIALS AND METHODS

Study area. The region of Kakheti is located in a southeast part of Georgia. The region is characterized sufficiently complex geotectonic structure. The territory were samples of rocks have been selected is located in the foothills of the Main Caucasian Range in the east part of Kakheti. The investigated area includes two tectonic zones (see Figure 1), in particular:

- Kazbek-Lagodekhi zone (folded-flaky) (I_2);
- Mestia-Tianeti zone (fold-napped) (I_3).

Twenty rock samples (Table 1) were collected in the investigated area (located in the canyon of the river Stori along the way from the settlement Pshaveli to the settlement Omalo, and also near settlements Lopota (Lp), Eniseli (En), Tsitskanaantseri (Tf), Mtisdziri (Mz), Lapniani (Ln), Baisubani (Bs), and cities of Kvareli (Kr) and Akhmeta (Ah)), in particular:

- in zone I_2: thirteen samples;
- in zone I_3: seven samples.

The types of samples collected were as following:

- igneous, two samples including:
  - effusive – two samples:
    - diabase – one sample (105);
    - basalt – one sample (111);
- sedimentary, eight samples including:
  - sandstone – six samples (109, 119, 122, 127, 132, 135);

- argillite – two samples (133, 137);
- metamorphic, ten samples including:
  - sedimentary, six samples including:
    - clay-shale – six samples (102, 103, 106, 108, 113, 129);
  - volcanic, four samples, including:
    - shale – three samples (98, 99, 101);
    - marble – one sample (116).

All selected samples are of the Jurassic period.

Figure 1 shows layout of locations, and Figure 2 and Figure 3 show locations Pv-8 and Kr-3, respectively.

Table 1 List of locations (L), sample numbers (SN), types (ST) of investigated samples

#	Tc	L	SN	Lt(N); Ln(E)	ST
1	I_2	Pv-1	98	42.22982; 45.48529	Shale (chlorite-sericite, greyish) [Sl (Chl,Sr,Gr)]
2	-“-	Pv-2	99	42.22980; 45.48534	Shale (chlorite-sericite, greyish) [Sl (Chl,Sr,Gr)]
3	-“-	Pv-4	101	42.22892; 45.48372	Shale (quartz-sericite) [Sl (Q-Sr)]
4	-“-	Pv-5	102	42.22855; 45.48335	Clay-shale (rust with quartz veins) [Cl-Sl (Q,Rt)]
5	-“-	Pv-7	104	42.22732; 45.48134	Clay-shale (quartz-chlorite-sericite, rust) [Cl-Sl (Q,Chl,Sr,Rt)]
6	-“-	Pv-8	105	42.22668; 45.48226	Diabase (with pyrite flecks) [Db (Pyr)]
7	-“-	Pv-9	106	42.20423; 45.46114	Clay-shale (greyish) [Cl-Sl (Gr)]
8	-“-	Pv-11	108	42.19192; 45.45644	Clay-shale (rust with quartz veins) [Cl-Sl (Q,Rt)]
9	-“-	Pv-12	109	42.19067; 45.44211	Sandstone (silt quartzose) [Ss (Slt,Q)]
10	-“-	Pv-14	111	42.18785; 45.44946	Basalt (greyish) [Bs (Gr)]
11	-“-	Pv-16	113	42.15329; 45.42042	Clay-shale (greyish) [Cl-Sl (Gr)]
12	-“-	Lp-5	119	42.06280; 45.53373	Sandstone (carbonate silt) [Ss (Cr, Slt)]
13	-“-	Lp-2	116	42.07293; 45.60978	Marble (white) [Mrb (W)]
14	I_3	Ah-2	122	42.05140; 45.24077	Sandstone marly with argillite [Ss (Mr,Ar)]
15	-“-	En-2	127	42.00269; 45.66016	Sandstone (carbonate) [Ss (Cr)]
16	-“-	Kr-3	129	41.99424; 45.84642	Clay-shale (argillaceous) [Cl-Sl (Ar)]
17	-“-	Tf-1	132	41.90741; 45.89193	Sandstone (silt, carbonate) [Ss (Slt, Cr)]
18	-“-	Mz-2	133	41.85249; 46.04052	Argillite (carbonate) [Ar (Cr)]
19	-“-	Ln-2	135	41.83746; 46.09737	Sandstone (silt) [Ss (Slt)]
20	-“-	Bs-2	137	41.82818; 46.18119	Argillite (black) [Ar (Bk)]

Notes: Tct – tectonic zone; Lt(N) – latitude (north); Ln(E) – longitude (east).

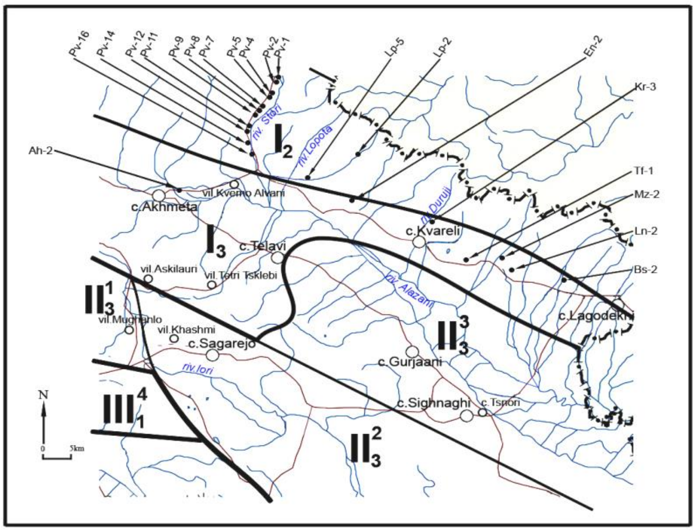

Figure 1. Layout of locations.

Figure 2. Location Pv-8. **Figure 3. Location Kr-3.**

Sampling and analysis

Sampling Samples were selected from the outcropped rocks and put in plastic containers (volume up to 2.0 L). After drying in the laboratory, samples were broken into pieces < 40 mm and were then crushed using a special crusher (jaw crusher Retsch) to a size of approximately 1 mm. Then samples were dried at 105 - 110°C to constant weight and their bulk density was then determined. These values were used for the description of sample geometry. The samples were sealed in Marinelli beaker and stored for more than four weeks to achieve secular equilibrium between Ra-226 and Rn-222.

Measurement of gamma radiation activit

Measurements were carried out using a gamma spectrometer Canberra GC2020 with a semi-conductor germanium detector with relative efficiency of 24%. Gamma spectra acquisition time was 72 hour. Genie-2000 S500 software with additional modules was used for the analysis, in particular, S506 Interactive Fit Program. By means of this program, for all spectra a “decomposition” of the interference peak in the area of 186 keV was carried out (program identifies one peak in this area that is growing out of an interference of two closely spaced peaks of U-235 (185.715 keV) and Ra-226 (186.211 keV)). The program S506C processes the spectral curve mathematically; therefore, in this area two peaks are created with energies corresponding to U-235 and Ra-226. During the program identification of peaks and calculation of activity concentration a tolerance value was established in such a manner that low-energy peak was compared only with U-235 and high-energy peak only with Ra-226. Results show that, in particular, determination uncertainty of the activity concentration of Ra-226 was within 11 and 40%. Its activity was compared with the activity of its daughters, Pb-214 and

Bi-214, which had a determination uncertainty between 2 and 4%. Values of Ra-226, Pb-214 and Bi-214 activity did not differ sufficiently. Thus, it is possible to consider that similar determination uncertainty of Ra-226 concentration is satisfactory; thus, this method was also used for the determination of U-235 activity concentration by the 185.715 keV line. Received values of U-235 activity were compared to values of U-238 activity (which were determined by the line 63.3 keV line of Th-234, with an uncertainty range of 6.6-8.7%). The value of their activities ratio U-238/U-235, which is considered as constant (21.7) for natural objects [10], was used as a criterion. On occasion, in low-activity samples, Ra-226 activity was specified by the average activity of Pb-214 and Bi-214 and obtained value was considered a definitive estimation of U-235 activity. For Th-232 activity determination average values for Ac-228, Ra-224, Pb-212, Bi-212, and Tl-208 were used, which had determination uncertainties limits between 2.1 and 11.7% (except for the sample 116 for whom value of determination uncertainty for radionuclide Bi-212 has made 81.3%). Activities ratios were also determined U-238/Th-232 (which is accepted as being equal 0.81 for the closed systems [11, 12], Ra-226/U-238 and Pb-210/Ra-226 (equilibrium value 1.00), which are used to estimate the mechanism of various geochemical processes.

Taking into account the influence of matrix composition, the chemical composition of samples was determined on the basis of literary data [13, 14], which were then used in the special software (LabSOCS) for efficiency calibration of the activity concentration calculation. System LabSOCS allows to create calibrations by laboratory quality efficiency without application of radioactive calibrate sources. For radionuclides identification a special library was used that contains lines of 41 radionuclides and other specific sources (in total 351 lines). Database NuDat [15] was used for library compiling. For activity (A) calculation, the background radiation was subtracted.

The assessment of values of radium equivalent activity Ra_{eq} (Bq/kg) was carried out using the formula [16]:

$$Ra_{eq} = A_U + 1.43 A_{Th} + 0.07 A_K$$

where A_U, A_{Th}, and A_K are the activity concentrations (Bq/kg) of U-238, Th-232 and K-40, respectively.

For samples characterization by radioactivity degree, taking into account accepted limit value of Ra_{eq} (370 Bk/kg; equivalent to the annual γ-radiations dose of 1.5 mSv/y) [17]) some groups were established according to their value of equivalent activity, in particular:

1st group: nonradioactive samples (activity is low and did not exceed 30 Bq/kg);

2nd group: samples with low radioactivity (activity is in the range of 30 to 100 Bq/kg);

3rd group: samples with average radioactivity (activity is in the range of 100 to 300 Bq/kg);

4th group: samples with high radioactivity (activity is in the range of 300 to 1000 Bq/kg).

The technique is described in more detail in works [8, 9].

RESULTS

Up to 22 radionuclides were identified from the results of analysis of the gamma spectra of rock samples: the Th-232 family (Ac-228, Th-228, Ra-224, Pb-212, Bi-212, Tl-208); the U-238 family (Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, Pb-210); the U-235 family (U-235, Th-231, Th-227, Ra-223, Rn-219, Pb-211); the natural radionuclides Be-7, K-40, the technogenic radionuclide Cs-137 (several specific lines were also identified that were incipient, as a result of the interaction of cosmic rays with the material of the detector or the sample).

The average activity of identified families of radionuclides varied widely, from 0.10 Bq/kg (for the U-235 family) to 98.6 Bq/kg (for the Th-232 family). Of the individual radionuclides, K-40 had the highest activity (up to 1475 Bq/kg). In some samples, the activity of radionuclides was lower than the Minimal Detectable Activity (MDA). The activity concentrations of the main radionuclides of the investigated samples, the equivalent activity, the activity ratios and their averages (av), the minimal (mn) and maximal (mx) values, among other data, are given in Tables 2 to 6. Figure 2 shows the statistically significant correlation between the U-238 and Th-232 activity concentration.

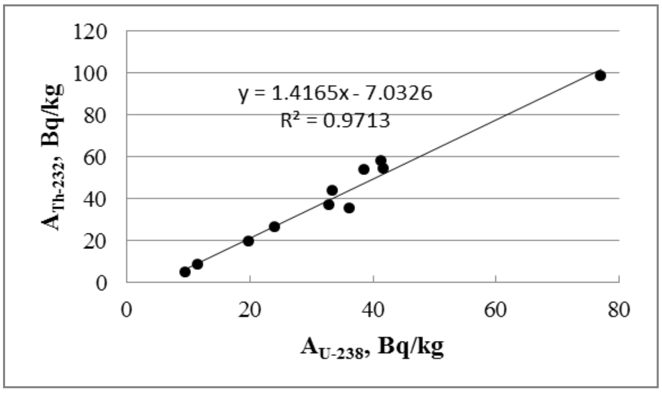

Figure 4. Correlation between U-238 and Th-232.

Table 2

The activity concentrations (A, Bq/kg) of the radionuclides of families Th-232, U-238 (Th-234), U-235, Ra-226, Pb-214, Bi-214, Pb-210, radionuclides Be-7, K-40 and Cs-137, the equivalent activity (Ra_{eq}, Bq/kg), the activity ratios U-238/U-235; U-238/Th-232, Ra-226/U-238 and Pb-210/Ra-226,

#	L	SN	A, Bq/.kg										Ra_{eq}	U-238/	U-238/	Ra-226/	Pb-210/
			Th-232	U-238	Ra-226	Pb-214	Bi-214	Pb-210	U-235	Be-7	K-40	Cs-137	Bq/kg	U-235	Th-232	U-238	Ra-226
1	Pv-1	98	5.2	9.6	8.3	9.0	8.0	23.0	0.45	<M	318	0.21	39.3	21.3	1.85	0.86	2.77
2	Pv-2	99	19.9	19.8	18.3	18.9	18.0	27.4	0.95	<M	557	0.42	87.2	20.8	0.99	0.92	1.50
3	Pv-4	101	26.3	24.0	27.4	24.2	23.6	19.6	1.09	—	490	0.61	95.9	22.0	0.91	1.14	0.72
4	Pv-5	102	98.6	77.0	82.3	78.8	75.4	62.5	3.53	—	1475	0.63	321	21.8	0.78	1.07	0.76
5	Pv-7	104	37.0	32.9	31.9	33.5	31.2	<M	1.52	—	1124	—	165	21.6	0.89	0.97	—
6	Pv-8	105	35.4	36.2	37.0	36.5	34.5	49.9	1.65	—	1172	0.70	169	21.9	1.02	1.02	1.35
7	Pv-9	106	54.1	38.6	44.0	39.3	38.7	38.2	1.72	—	802	—	172	22.4	0.71	1.14	0.87
8	Pv-11	108	43.8	33.4	33.6	32.2	31.3	26.3	1.51	—	156	—	107	22.1	0.76	1.01	0.78
9	Pv-12	109	54.5	41.8	33.6	37.0	36.2	60.4	2.00	<M	827	—	178	20.9	0.77	0.80	1.80
10	Pv-14	111	1.11	<M	4.2	2.5	2.6	19.0	0.10	—	21	—	-	—	—	—	4.52
11	Pv-16	113	58.0	41.4	41.5	40.7	39.4	—	1.90	—	766	<M	178	21.8	0.71	1.00	—
12	Lp-5	119	8.7	11.6	13.3	11.7	11.0	17.3	0.53	<M	95	0.22	30.7	21.9	1.33	1.15	1.30
13	Lp-2	116	0.61	<M	3.0	2.5	2.3	13.0	0.14	—	4.5	—	—	—	—	—	4.33
14	Ah-2	122	9.7	11.2	15.4	12.0	11.8	12.9	0.53	—	173	0.20	37.2	21.1	1.15	1.38	0.84
15	En-2	127	4.7	9.1	12.0	10.2	9.9	18.5	0.42	<M	52	0.51	19.5	21.7	1.94	1.32	1.54
16	Kr-3	129	52.8	39.8	33.7	35.5	34.3	35.0	1.90	—	710	1.2	165	20.9	0.75	0.85	1.04
17	Tf-1	132	41.0	29.5	26.9	26.8	26.3	39.8	1.38	—	709	0.32	138	21.4	0.72	0.91	1.48
18	Mz-2	133	18.7	18.5	12.5	15.1	14.4	16.4	0.90	—	319	<M	67.6	20.6	0.99	0.68	1.31
19	Ln-2	135	30.3	25.5	24.3	25.0	24.0	30.1	1.17	—	376	0.10	95.1	21.8	0.84	0.95	1.24
20	Bs-2	137	50.5	33.3	40.7	36.0	35.1	48.3	1.53	—	778	—	160	21.8	0.66	1.22	1.19
		av	*32.5*	*29.6*	*27.1*	*26.4*	*25.4*	*31.0*	*1.21*	—	*546*	*0.47*	*124*	*21.6*	0.99	1.02	1.63
		mn	*0.61*	*9.1*	*3.0*	*2.5*	*2.3*	*12.9*	*0.10*	—	*4.5*	*0.10*	*19.5*	*20.6*	0.66	0.68	0.72
		mx	*98.6*	*77.0*	*82.3*	*78.8*	*75.4*	*62.5*	*3.5*	—	*1475*	*1.2*	*321*	*22.4*	1.94	1.38	4.52

Table 3

Generalized data – the activity concentration of radionuclides (A, Bq/kg) of families (Th-232, U-238, U-235) and radionuclide K-40, the equivalent activity (Ra_{eq}, Bq/kg) depending on sample type (G)

G	**GR**	**A, Bq/kg**					***Ra*$_{eq}$*,***	**U-238/**	**Ra-226/**	**Pb-210/**
		Th-232	**U-238**	**Ra-226**	**U-235**	**K-40**	**Bq/kg**	**Th-232**	**U-238**	**Ra-226**
Ig	*Ef*	18.3	36.2	20.6	0.88	597	169	1.02	1.02	2.94
Sd		29.7	23.7	23.8	1.11	460	98.4	0.97	1.02	1.31
	Ss	24.8	21.5	20.9	1.01	372	83.0	1.13	1.08	1.37
	Ar	34.6	25.9	26.6	1.22	549	114	0.82	0.95	1.25
Mt		35.2	30.8	29.4	1.34	591	129	1.01	0.99	1.60
	Sd	57.4	43.9	44.5	2.01	839	185	0.77	1.01	0.86
	Vc	13.0	17.8	14.3	0.66	342	74.2	1.25	0.98	2.33

Note. GR – group of rocks.

Table 4

Distribution of the average values $Ra_{eq\text{-}av}$ of the equivalent activity Ra_{eq} by the activity level group (GA), their quantity (N_s) and percentage (r, %).

#	**GA**	***Ra*$_{eq}$, Bq/kg**	***Ra*$_{eq\text{-}av}$, Bq/kg**	**N_s**	**r, %**
1	I	<30	19.5	1	5.6
2	II	30-100	64.7	7	38.8
3	III	100-300	159	9	50.0
4	IV	300-1000	321	1	5.6

Table 5

Generalized data – the activity concentration of radionuclides (A, Bq/kg) of families (Th-232, U-238, U-235) and radionuclide K-40, the equivalent activity (Ra_{eq}, Bq/kg) depending on the type of tectonic zone (Tct)

Tct	A, Bq/kg					*Ra*$_{eq}$,	U-238/	Ra-226/	Pb-210/
	Th-232	U-238	Ra-226	U-235	K-40	Bq/kg	Th-232	U-238	Ra-210
I_2	34.1	33.3	29.1	1.31	601	140	0.98	1.01	1.88
I_3	29.7	23.8	23.6	1.11	445	97.4	1.01	1.04	1.23

General characteristics

The activity of the families of radionuclides varied in the samples by more than two orders of magnitude (Table 2); in particular, the activity of Th-232 varied from 0.61 to 98.6 Bq/kg (average value of 32.5 Bq kg-1), U-238 – from 9.1 Bq/kg to 77.0 Bq/kg (average value of 29.6 Bq/kg); and U-235 – from 0.10 to 3.5 Bq/kg (average value of 1.21 Bq/kg). The activity of K-40 varied by more than two orders of magnitude, from 4.5 to 1475 Bq/kg (average value of 546 Bq/kg). Be-7 was found in trace amounts in some samples. The technogenic radionuclide Cs-137 was measured in eleven samples in small amounts (0.10–1.2 Bq/kg), and was found in some samples in trace amounts. For the activity ratio of U-238/U-235, all the obtained values correspond (within ±10%) to a value of 21.7 (accepted for natural objects). The activity ratio of U-238/Th-232 showed marked deviation (more than ±10%) from the average value of 0.81 (for closed systems); an increase was observed in nine samples (ranging from 0.89 to 1.94), and a decrease in four samples (0.66–0.72). The value of the ratio Ra-226/U-238 differs (by more than ±10%) from the equilibrium value; in six samples, it is greater than equilibrium (1.14–1.38), and in four samples it is less (0.68–0.86). The ratio Pb-210/Ra-226 also differs from the equilibrium value (by more than ±20%[1]) in some samples; in eleven samples, it is greater than equilibrium (1.24–4.52) and in three samples it is less (0.72–0.78). (Note: activity ratios were not determined for all samples, because in some samples the activities of the corresponding radionuclides were below the MDA or were not measured). Within the Th-232 – Tl-208 chain, equilibrium was basically observed (except for Th-228, for which the determination of uncertainty was appreciable more than for other radionuclides). The majority of samples (88.8 %) by the level of the radium equivalent activity have low and average radioactivity, and the smallest quantity of samples (5.6%) applies to the group of nonradioactive samples as well as to the group with high radioactivity – 321 Bq/kg (see Table 6).

Dependence on the Type

The average activity of naturally occurring radionuclides in igneous rocks (was calculated only for one type – diabase) is greater (Table 3) than in metamorphic and sedimentary rocks (the average activity 169, 129, and 98.4 Bq/kg, respectively). The highest activity was found for the group of clay-shale (185 Bq/kg) that is connected with rather high content of radionuclide K-40 (the maximum value among all investigated samples - 1475 Bq/kg), and also Th-232 (57.4 Bq/kg) and U-238 (43.9 Bq/kg).

The calculated values of the ratios U-238/Th-232 for samples of sedimentary and metamorphic rocks are about of the same level – from 0.66 to 1.94 (the average value of 0.97) and from 0.71 to 1.85 (the average value of 1.01), at the same time the greatest diapason was observed for sandstones (0.72-1.94), and least – for the group of clay-shales (0.71-0.89). The ratio Ra-226/U-238 for samples of sedimentary rocks varied within a wide range (from 0.68 to 1.38) in comparison of metamorphic rocks (0.85-1.14) at the same time the greatest diapason was observed for argillites (0.68-1.22), and the least for clay-shales (0.85-1.14).

Dependence on the tectonic zones.

The average equivalent activity of the studied samples selected in the zone $\mathbf{I_3}$ (97.4 Bq/kg) is rather smaller than in the zone $\mathbf{I_2}$ (140 Bq/kg), however activity of several samples (clay-shale, sample 129) reach higher values (165 Bq/kg). Values of activity ratios for two zones are comparable among themselves (Table 5).

[1] The range of limits is expanded, because determination uncertainty of Pb-210 reached up to 20%.

DISCUSSION

The concentration of radioactive elements in rocks and soils is formed by the radioactivity of original structures and the whole set of subsequent processes of rock and soil formation. The content and concentration of NORM identified in the investigated samples generally correspond to those observed [18] for various rocks and soils. For the study region, this is the first time such analysis has been carried out. All these radionuclides, except Cs-137, are of natural origin. They are also characteristic for the region of Georgia [8, 9].

Rock samples were selected in a geological area characterized by a sufficiently complex geotectonic structure. Basically, types of the selected samples corresponded to sedimentary and metamorphic groups (besides there were selected two igneous samples). Each of these groups has specific mineralogical and chemical composition as well as rock-forming mechanism, which is connected to the wide range of radioactivity concentration values, taking place practically for all identified radionuclides, and also for activities ratios of some radionuclides.

As it has been noted above, the greatest radioactivity among the investigated samples have the clay-shales concerning to metamorphic sedimentary rocks. It is necessary to notice, that sedimentary rocks, including metamorphic rocks of sedimentary origin, inherit a radioactivity of rocks from which they are formed. According to literary data, the highest and rather constant radioactivity among sedimentary rocks have clay-shales (and clays too). Relatively high radioactivity of clay-shales is explained by both the raised sorption of uranium, radium, thorium, and potassium from the natural fluids on the clay particles and relatively high potassium content in these rocks (up to 6.5 %).

In all samples the U-238/U-235 ratios observed correspond, within error, to the natural value, that, besides methodological aspect, allows making the conclusion about absence of pollution by anthropogenous U-235. The results show a deviation of the ratio U-238/Th-232 from the average value, which may be a consequence of the fact that the given system was essentially closed. The raised values of the ratio Ra-226/U-238 noted also for much samples, apparently, are connected with prevalence of dissolution processes of U-238 (that leads to decrease of its concentration), and lowered values are connected with prevalence of leaching processes of Ra-226 from surface strata (where samples were selected). The deviation of ratio Pb-210/Ra-226 from the average value is not characteristic for rocks in connection with insignificant migration of radon in relative solid rocks and insignificant effect of accumulation of atmospheric Pb-210 on a surface of rocks (because of washing off by atmospheric precipitates).

In some samples an insignificant concentration of naturally occurring radionuclide Be-7 (a so-called cosmogeneous radionuclide that is formed as a result of nuclear reactions in an upper atmosphere) are observed as a result of precipitation and in some cases can be identified in gamma-spectra.

The technogenic radionuclide Cs-137 also gets into the samples as a result of atmospheric precipitation. Usually, the presence of Cs-137 in natural objects (for example, in soil) is linked to the Chernobyl disaster. After that it decreased in quantity as a result of decay and migration. In several samples an insignificant concentration (in comparison with soils where its concentration is considerably higher) was observed that, apparently, is connected with intensive process of washing from a sample surface.

Table 6 shows some reference data in other regions of the world. Apparently, the values received in the present work, on the average, are comparable with data of other regions.

Table 6

Comparison of radioactivity of rock samples with other areas of the world.

Country	A, Bq/kg			Ref.
	U-238	Th-232	K-40	
Brazil	31	73	1648	[19]
Cyprus	1-588	1-906	50-1606	[20]
Albania	8-27	13-40	266-675	[21]
India	0.44-50.83	0.21-293.67	233.49-2091.70	[22]
Turkey	13.80-29.25	26.12-44.39	307.54-539.14	[1]
Italy	42-70	31-37	410-475	[23]
Greece	29-110	19-88	152-1593	[24]
Algeria	11-25	6-32	56-607	[25]
Georgia	9.1-77.0	0.61-98.6	4.5-1475	This study

In conclusion it is necessary to notice, that the received results represent doubtless scientific and applied interest for investigated region that confirms an urgency of such researches and necessity of their systematic character.

CONCLUSION

1. It was established that in rock samples there are up to 22 detected radionuclides, in particulat, the radionuclides of families Th-232, U-238, U-235, other naturally occurring radionuclides – Be-7**,** K-40, and the technogenic radionuclide Cs-137.
2. The main features and regularities of samples radioactivity were established, in particular:
   - activity of families radionuclides and the radionuclide K-40 varied in various samples by more than two orders of magnitude – from 0.61 to 1475 Bq/kg; the U-238/U-235 activity ratio corresponds to the value of 21.7 (accepted for natural objects); U-238/Th-232 ratio deviations (more than ±10%) from the average value of 0.81 (for closed systems) were observed as both increases and decreases; deviations of ratios Ra-226/U-238 and Pb-210/Ra-226 from the equilibrium value (1.0) were insignificant;
   - radionuclide Be-7 was measured in some samples at trace amounts;
   - the technogenic radionuclide Cs-137 was measured in some samples (activity concentration from 0.10 to 1.2 Bq/kg).
3. Some features of activity distribution were determined depending on sample type and depending on geotectonic zones.
4. Analysis of obtained results and some of their features was carried out, as well as comparison with literary data.

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