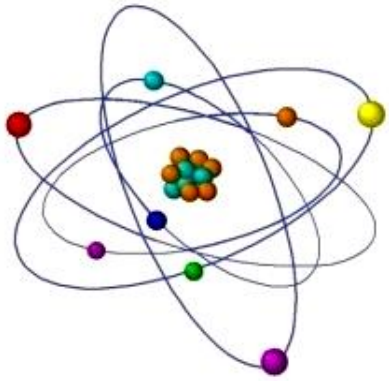
STUDIES OF NATURAL RADIOACTIVITY IN ROCKS OF SOUTH CAUCASUS
(Kazbegi region, Georgia)



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ABSTRACT: *There are given results of radioactivity research in the rock samples within the territory of Kazbegi municipality, Georgia. Twenty-three samples have been collected from 22 locations. Up to 22 radionuclides were identified. Activity concentration of radionuclides of Th-232 family was in the limits from 19.2 to 57.9 Bq/kg (mean, 42.4 Bq/kg), U-238 family – from 16.5 to 38.5 Bq/kg (mean, 31.1 Bq/kg), U-235 family - from 1.03 to 2.40 Bq/kg (mean, 1.91 Bq/kg). Also, individual radionuclides have been identified: K-40 – from 204 to 1069 Bq/kg (mean, 632 Bq/kg), Cs-137 – from 0.18 to 2.69 Bq/kg. Radium equivalent activity varied from 58.2 to 192 Bq/kg (mean, 136 Bq/kg).*

Key words: radionuclides, rocks, activity concentration, South Caucasus

INTRODUCTION

Radioactivity is a part of the natural environment: the earth's surface, rocks, atmosphere, human body, food. The main part of the Earth’s radioactive elements is contained in the rocks that make up the Earth's crust. From here, radioactive elements pass into the soil, then into plants, and finally, together with plants, they enter animal and human organisms.

The radioactivity of rocks and ores is determined by the content of radioactive rock-forming minerals. Depending on the qualitative and quantitative composition of these minerals (in particular, uranium-containing rocks), the conditions of formation, age and degree of metamorphism, their radioactivity varies very widely. The radioactivity of rocks and ores is higher when the concentration of minerals containing naturally occurring radioactive elements (mainly the families of uranium, thorium, and potassium-40) is higher.

The radioactive elements of various radionuclides in rocks may affect the health. Thus, numerous studies have been performed in many regions of the world, and the obtained data can be used to establish if the local controls are needed.

For example, previous work [1] has focused on samples of igneous rocks (dolerite, granite, and granitic gneisses), sedimentary rocks (shales, limestones, sandstones) and metamorphic rocks (black shale, slate, graphite, marble, quartz mica, calcareous schist, and quartz), selected in the state of Kashmir (Pakistan). As a result of this work it was determined that activity

concentration of Ra-226 varied in the range from 4.63 to 108.54 Bq/kg (mean activity concentration was 37.32 Bq/kg), the concentration of Th-232 in the range of ≤1.80 – 110.73 Bq/kg (mean activity - 38.57 Bq/kg), and the concentration of K–40 - in the range of 9.55 – 1135.55 Bq/kg (mean activity – 465.62 Bq/kg). The authors noted that the concentrations of Cs-137 activity in all samples were below the detection limit. The radium equivalent activity of the samples was estimated, which varied in the range of 20.6 - 294 Bq/kg (the average activity value was 126 Bq/kg), which is significantly lower than the maximum recommended value of 370 Bq/kg [2, 3].

In a different work [4], samples of Tufa deposits from four separate occurrences in the Dinaric Region of Croatia were studied. It was found that the activity concentration of Th-232 varied in the range of 1.0 - 12.9 Bq/kg with an mean value of 4.5 Bq/kg; the mean average concentration of U-238 was noticeably higher and amounted to 7.5 Bq/kg (range 1.3 - 14.6 Bq/kg); results comparable to U-238 were obtained for Ra-226 – the mean value was 7.3 Bq/kg, the range is from 2.9 to 13.0 Bq/kg. The technogenic radionuclide Cs-137 was also found in the studied samples, which activity concentration varied in the range of 3.9 - 77.8 Bq/kg with mean value of 24.6 Bq/kg; the authors explain this as contamination of Cs-137 originating from rainwater; obsolete calcareous tuff is not a closed system, and sampling was performed after an accident in 1986, which was accompanied by heavy rains in this area. The ratios of radionuclide activities were also calculated, the values of which for U-238/Th-232 varied in the range of 0.81 - 5.30 with the mean value of 2.41, and for Ra-226/U-238 in the range of 0.59– 2.22 with the mean value of 1.10.

Studies of various rocks used as building materials have been conducted in Romania [5]. The activity concentrations of the radionuclides Th-232, Ra-226, K-40, and Cs-137 in tufa were 9.8, 10.5, 180, and 9.6 Bq/kg, respectively.

Authors of the work [6] examined several dozen limestones samples, selected throughout Turkey. The activity concentration of Ra-226 varied in the range of 0.7 - 55.1 Bq/kg with the mean value of 19 Bq/kg, Th-232 – in the range of 1.2 - 20.9 Bq/kg with the mean value of 4.3 Bqkg, and K-40 – in the range of 10.1 - 258.4 Bqkg with the mean value of 55 Bq/kg. Based on the measurement results, the equivalent activity values were calculated, which varied in the range of 3.2 - 81.6 Bq/kg with the mean value of 29.4 Bq/kg.

Studies of the radioactivity of various environmental objects in Georgia have been conducted in the past and were mainly stimulated by the accident at the Chernobyl nuclear power plant in 1986.

The authors of this work participated in a number of studies of radioactivity on the territory of Georgia, including the radioactivity of rocks [7, 8, 9, 10, 11].

In this work, a study of natural and technogenic radioactivity was conducted in the part of the territory of the Kazbegi municipality. The aim of the work was to establish the radionuclide content and background concentrations of radionuclides in various geological structures in the study area, to study the level of gamma-ray radioactivity, as well as to assess the corresponding radiological risk to the local population and tourists.

MATERIALS AND METHODS

Study area:

Kazbegi municipality is part of the Mtskheta-Mtianeti region. The municipality is located entirely in the mountainous part of Georgia. The height above sea level varies from 1,700 m to 5,000 m. The area of the municipality is 1081.7 km^2. The municipality borders the Russian Federation in the northern part.

The main orographic unit of the territory is the Main Caucasian Ridge. The relief of the municipality is mountainous in nature – mostly rocky and difficult to access. Erosive, volcanic and glacial landforms are developed. Karst rocks are found sporadically. The basis of the Kazbegi mountain range consists of sandstones that formed in the Jurassic period, as well as basalt formations such as quartzite, clay shales and clay deposits. Lava pillows are a part of the geology of the region. Cliffs have formed on the slopes of the Daryal Gorge, on the walls of which basalt sections and layers of frozen lava are visible.

Twenty-three rock samples were selected at twenty-two control points in the study area (two samples of different rocks were selected at the control point Kz-33). Table 1 contains a list of control points, their brief description, geographical coordinates, as well as a list of selected samples and their types.

Sampling and analysis:

Sampling and sample preparation

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 then were 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 activity

Measurements of the radionuclide content of the studied samples and the activity concentration of radionuclides were carried out using a Canberra GC2020 gamma-spectrometer with a semiconductor germanium detector with a relative efficiency of 24%. The acquisition time for gamma spectra was 48 hours. The Genie-2000 S500 software with additional modules was used for the analysis, in particular, S506, an Interactive Fit Program. This program was used to “decompose” the interference peak in the 186 keV region in all spectra (in this region, the program identifies one peak resulting from the interference of two closely spaced peaks U-235 (185.715 keV) and Ra-226 (186.211 keV)). Using the S506C program, the spectral curve is mathematically processed, as a result of which two Gaussian peaks with energies corresponding to U-235 and Ra-226 are created in this region. When programmatically identifying peaks and calculating the concentration of activity, the tolerance value was set in such a way that only U-235 was compared to the low–energy peak, and only Ra-226 to the high-energy peak. As the results showed, in particular for Ra-226, the error in determining its activity concentration was mainly in the range of 8.8 - 24%. His activity

was compared with the activity of his daughters Pb–214 and Bi-214, the error in determining which lay in the range from 2.4 to 3.0%. The obtained activity values of Ra-226, Pb-214 and Bi-214 differed little from each other. Thus, it can be considered that the error in determining the concentration of Ra-226 in this way is quite satisfactory, and this method was also used to determine the activity concentration of U-235 along the 185.715 keV line. The obtained values of U-235 activity were compared with the values of U-238 activity (which was determined by the Th-234 - 63.3 keV line with an error in the range of 6.3 - 14%). The value of the ratio of their activities (U-238/U-235 = 21.7 [12]) was used as a criterion. With large deviations from this value (more than 10%), the 185.715 keV line (as well as the 63.3 keV line) was reanalyzed using the S506 program. (In some cases, in low-activity samples, the activity of Ra-226 was refined by the average activity of Pb-214 and Bi-214 and the resulting value was taken into account in the final assessment activity values U-235). To determine the activity of Th-232, the average values for Ac-228, Ra-224, Pb-212, Bi-212 were used, the errors of which ranged from 3.2 to 7.2%. The activity ratios U-238/Th-232 (which for closed systems is assumed to be 0.81 [13, 14]), Ra-226/U-238 and Pb-210/Ra-226 (equilibrium value 1.00) was also determined, which are used to evaluate the mechanism of various geochemical processes.

Table 1
List of control points (CP), types (ST) of investigated samples

#	CP	Lt(N); Ln(E)	Description of CP	SN	ST
1	Kz-1	42.7332; 44.6319	Mtavarangelozi Monastery complex	416-R	St
2	Kz-4	42.7351; 44.6353	“-“	401-R	Dt
3	Kz-5	42.6670; 44.6299	Stepantsminda-Larsi road, Chkhere River gorge	398-R	An
4	Kz-10	42.6937; 44.6410	Terek River valley	415-R	St
5	Kz-20	42.6420; 44.6364	Tbilisi-Stepantsminda road, near the entrance to Stepantsminda	414-R	St
6	Kz-21	42.6381; 44.6352	Near the confluence of the rivers Terek and Snostskali	399-R	Cl-Sl
7	Kz-28	42.5595; 44.7022	Villagrs Karkucha-Juta road	409-R	Sl
8	Kz-31	42.5948; 44.6697	Village Akhaltsikhe	413-R	Al
9	Kz-33	42.6035; 44.5682	Village Goristsikhe	418-R	An-Bs
10	“-“	“-“	“-“	420-R	An
11	Kz-37	42.5719; 44.5240	Village Kobi	417-R	An
12	Kz-39	42.5604; 44.5121	Village Kobi	402-R	An
13	Kz-42	42.5849; 44.5535	Village Kanobi	403-R	Tf
14	Kz-50	42.5516; 44.4944	Surroundings of Kobi village	406-R	Ss
15	Kz-54	42.5380; 44.4763	Cross Pass Travertine	411-R	Dc
16	Kz-55	42.6236; 44.6097	Near the entrance to the village Arsha	397-R	Cl-Sl
17	Kz-59	42. 6230; 44.5983	Village Arsha, Gaiboteni	405-R	An
18	Kz-61	42.6237; 44.6098	Surroundings of Arsha village	404-R	Cl-Sl
19	Kz-64	42.6099; 44.5740	Village Tkarsheti	400-R	An
20	Kz-66	42.6582; 44.6229	Gergeti Trinity Church	412-R	Cl-Sl
21	Kz-69	42.6575; 44.6376	Village Gergeti	410-R	Cl-Sl
22	Kz-75	42.5000; 44.4836	Panorama Gudauri	407-R	Tf
23	Kz-76	42.4766; 44.4777	Village Gudauri	408-R	Tf

Notes: **1**. Lt(N) – latitude (north); Ln(E) – longitude (east). **2**. SN – sample number. **3.** *Rock types*: An - Andesite; Dc - Dacite; Dt - Diorite An-Bs - Andesite-basalt; Tf – Tuff; Ss - Sandstone; Al - Aleuropelite St – siltstone; Cl-Sl - Clay-slate; Sl – Slate.

To account for the effect of the matrix content, based on the literature data, the chemical content of rocks [15] was established, which were then used in special software (LabSOCS) for efficiency calibration in calculating the activity concentration. The LabSOCS system allows

you to create calibrations based on the effectiveness of laboratory quality without the use of radioactive calibration sources. A special library containing lines of 41 radionuclides and other specific sources (351 lines in total) was used to identify radionuclides. The NuDat database was used to compile the library [16].

To assess the potential radiation risk of terrestrial radioactivity for the population living in the studied region, various indicators were calculated.

To assess the radiological hazard, the radium equivalent activity of Ra_{eq} (Bq/kg) and the annual effective dose of AEDE (mSv/y) were calculated; the calculation was carried out according to the formulas [17]:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.07A_K \quad (1)$$

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

$$AEDE = D \times N_h \times k_1 \times k_2 \quad (2)$$

where N_h is the number of hours per year (=8760 h), k_1 is the conversion factor of the effective dose rate into the absorbed dose rate in the air for adults, 0.7 Sv/Gy, k_2 is the outdoor coefficient (the proportion of time spent outdoors) which is 0.2, D is the absorbed dose rate (nGy/h):

$$D = k_U A_U + k_{Th} A_{Th} + k_K A_K \quad (3)$$

where k_U, k_{Th}, k_K are the so–called dose coefficients equal to 0.462, 0.604 and 0.0417, respectively.

To characterize the samples according to the degree of radioactivity, taking into account the accepted limit value of Ra_{eq} - 370 Bq/kg (equivalent to a dose of gamma radiation of 1.5 μSv/y) [18] several groups of samples were identified in terms of equivalent activity, in particular:

- group 1 – non-radioactive samples with an activity of no more than 30 Bq/kg;
- group 2 – with low radioactivity in the range from 30 to 100 Bq/kg;
- group 3 – with average radioactivity in the range from 100 to 300 Bq/kg;
- group 4 – samples with increased radioactivity in the range from 300 to 1000 Bq/kg.

Calculations of other indicators were also carried out to assess the radiation hazard, in particular:

- External hazard index (H_{ex}) is a widely used hazard index which represents the external exposure, and который рассчитывался по формуле:
$$H_{ex} = (A_U/370) + (A_{Th}/259) + (A_K/4810) \quad (4)$$
- Internal hazard index (H_{in}): in addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs; the internal exposure to radon and its daughter progenies is quantified by the internal hazard index H_{in}, which is given by the equation:
$$H_{in} = (A_U/185) + (A_{Th}/259) + (A_K/4810) \quad (5)$$

The values of the indices (H_{ex}, H_{in}) must be less than unity for the radiation hazard to be negligible [19].

RESULTS AND DISCUSSION

According to the results of the analysis of gamma spectra up to 22 radionuclides are identified in rock samples - family Th–232 (Ac-228, Th-228, Ra-224, Pb-212, Bi-212, Tl-208), family U-238 (Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, Pb-210), family U-235 (U-235, Th-231, Th-227, Ra-223, Rn-219, Pb-211), separate radionuclides Be-7, K-40, Cs-137 (also some specific lines are identified that arise as a result of the interaction of cosmic rays with the material of detector or sample).

Table 2 and Table 3 show the results of measuring the activity concentration (A, Bq/kg) of the main radionuclides for the studied samples, radium equivalent activity (Ra_{eq}, Bq/kg), activity ratios, their average (av), minimum (mn) and maximum (mx) values, as well as other data.

Table 2
Activity concentration (A, Bq/kg) of radionuclides of the 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, as well as their average (av), minimum (mn) and maximum (mx) values in rocks of various types

#	FN	CP	ST	A, Bq/kg									
				Th-232	U-238	Ra-226	Pb-214	Bi-214	Pb-210	U-235	Be-7	K-40	Cs-137
				Magmatic									
1	398-R	Kz-5	An	31.3	26.8	20.5	25.9	24.3	<M	2.01	-	450	<M
2	400-R	Kz-64	An	41.0	34.5	35.0	32.5	29.6	34.2	2.15	-	610	<M
3	401-R	Kz-4	Dt	45.0	31.8	31.2	31.7	29.5	-	1.51	-	788	<M
4	402-R	Kz-39	An	38.2	29.3	21.7	30.3	28.3	<M	1.98	-	530	-
5	405-R	Kz-59	An	40.0	30.0	21.8	30.7	28.3	28.9	2.17	<M	522	0.54
6	411-R	Kz-54	Dc	37.6	25.8	18.5	26.7	25.7	<M	1.60	4.70	534	-
7	417-R	Kz-37	An	37.5	33.5	35.4	29.6	27.7	<M	2.30	5.58	526	<M
8	418-R	Kz-33	An-Bs	39.5	32.0	29.1	32.0	29.4	41.5	2.40	3.00	593	0.82
9	420-R	Kz-36	An	39.9	33.6	31.7	31.1	29.2	32.5	2.27	6.26	588	<M
			av	38.9	30.8	27.2	30.1	28.0	34.3	2.04	4.88	571	0.68
			mn	31.3	25.8	18.5	25.9	24.3	28.9	1.51	3.00	450	0.54
			mx	45.0	34.5	35.4	32.5	29.6	41.5	2.40	6.26	788	0.82
				Sedimentary									
10	403-R	Kz-42	Tf	42.5	31.5	25.5	33.6	31.2	24.0	2.23	<M	573	<M
11	406-R	Kz-50	Ss	42.4	31.1	27.8	30.3	28.2	30.9	1.91	4.17	632	0.49
12	407-R	Kz-75	Tf	28.1	21.6	24.7	21.1	19.5	24.4	1.21	4.23	441	1.46
13	408-R	Kz-76	Tf	31.7	24.4	20.1	25.5	23.3	20.4	1.49	<M	414	-
14	413-R	Kz-31	Al	57.9	34.5	34.1	33.4	31.4	34.4	2.20	6.63	911	<M
15	414-R	Kz-20	St	41.9	31.7	31.2	30.3	28.7	40.6	2.04	6.22	579	2.69
16	415-R	Kz-10	St	55.0	38.5	34.4	37.4	34.8	50.1	2.15	-	1069	<M
17	416-R	Kz-1	St	19.2	16.5	11.8	17.4	16.1	32.6	1.03	<M	204	-
			av	39.8	28.7	26.2	28.6	26.7	32.2	1.78	5.31	603	1.54
			mn	19.2	16.5	11.8	17.4	16.1	20.4	1.03	4.17	204	0.49
			mx	57.9	38.5	34.4	37.4	34.8	50.1	2.23	6.63	1069	2.69
				Metamorphic									
18	397-R	Kz-55	Cl-Sl	51.7	34.3	-	31.9	29.3	34.9	1.16	<M	844	1.03
19	399-R	Kz-21	Cl-Sl	43.5	36.5	33.2	33.8	31.4	29.1	1.83	1.82	605	<M
20	404-R	Kz-61	Cl-Sl	55.5	32.3	23.3	32.2	30.1	<M	1.96	3.48	910	0.18
21	409-R	Kz-28	Sl	52.4	35.9	39.6	33.1	31.0	21.1	2.12	10.7	703	0.40
22	410-R	Kz-69	Cl-Sl	50.7	35.0	26.7	34.1	31.8	39.0	2.04	<M	711	<M
23	412-R	Kz-66	Cl-Sl	52.0	34.7	34.7	32.5	30.6	-	2.10	7.13	800	0.86
			av	51.0	34.8	31.5	32.9	30.7	31.0	1.87	5.79	762	0.62
			mn	43.5	32.3	23.3	31.9	29.3	21.1	1.16	1.82	605	0.18
			mx	55.5	36.5	39.6	34.1	31.8	39.0	2.12	10.7	910	1.03

The activity of radionuclides of the families varies significantly in different samples (Table 2), in particular, Th-232 – in the range of 19.2-57.9 Bq/kg (on average 42.4 Bq/kg), U-238 – 16.5-38.5 Bq/kg (31.1 Bq/kg), Ra-226 - 11.8-39.6 Bq/kg (27.8 Bq/kg), U-235 - 1.03-2.40 Bq/kg (1.91 Bq/kg). K-40 activity varies in the range of 204-1069 Bq/kg (632 Bq/kg). Be-7 was recorded in 12 samples in the range from 1.82 to 10.7 Bq/kg (in some samples - in trace amounts) Technogenic radionuclide Cs-137 was recorded in 9 samples in small quantities (0.18-2.69 Bq/kg), and in some samples it was recorded in trace amounts. The activity ratios of U-238/U-235 vary in the range from 14.1 to 36.1 with an average value of 20.8, and generally correspond to the value of 21.7 (accepted for natural objects).

The ratio U-238/Th-232 in most samples (14 out of 23) is within ± 10% of the average value of 0.81 (for closed systems), and in the remaining samples deviate from the average value in a smaller direction (from 0.58 to 0.71).

Table 3

Radium equivalent activity (Ra_{eq}, Bq/kg), activity ratios U-238/U-235; U-238/U-235, Ra-226/U-238 and Pb-210/Ra-226, dose rate (D, nGy/h) and annual effective dose (AEDE, mSv/y) as well as their average (av), minimum (mn) and maximum (mx) values in rocks of various types

#	FN	CP	ST	Ra_{eq}, Bq/kg	D, nGy/h	AEDE, mSv/y	H_{ex}	H_{in}	U-238/ U-235	U-238/ Th-232	Ra-226 /U-238	Pb-210 /Ra-226
						Magmatic						
1	398-R	Kz-5	An	103	50.0	0.061	0.29	0.36	21.2	0.86	0.77	-
2	400-R	Kz-64	An	136	66.1	0.081	0.38	0.47	17.4	0.84	1.01	0.98
3	401-R	Kz-4	Dt	151	74.7	0.092	0.42	0.51	22.7	0.71	0.98	-
4	402-R	Kz-39	An	121	58.8	0.072	0.34	0.42	21.6	0.77	0.74	-
5	405-R	Kz-59	An	124	59.8	0.073	0.34	0.43	14.1	0.75	0.73	1.32
6	411-R	Kz-54	Dc	117	56.9	0.070	0.33	0.40	18.7	0.68	0.72	-
7	417-R	Kz-37	An	124	60.1	0.074	0.34	0.44	28.5	0.89	1.06	-
8	418-R	Kz-33	An-Bs	130	63.3	0.078	0.36	0.45	15.1	0.81	0.91	1.42
9	420-R	Kz-36	An	132	64.2	0.079	0.37	0.46	23.1	0.84	0.94	1.02
			av	***126***	**61.5**	**0.075**	**0.35**	**0.44**	***20.3***	***0.80***	***0.87***	***1.19***
			mn	***103***	**50.0**	**0.061**	**0.29**	**0.36**	***14.1***	***0.68***	***0.72***	***0.98***
			mx	***151***	**74.7**	**0.092**	**0.42**	**0.51**	***28.5***	***0.89***	***1.06***	***1.42***
						Sedimentary						
10	403-R	Kz-42	Tf	132	64.1	0.079	0.37	0.45	20.9	0.74	0.81	0.94
11	406-R	Kz-50	Ss	136	66.3	0.081	0.38	0.46	20.8	0.73	0.89	1.11
12	407-R	Kz-75	Tf	92.6	45.3	0.056	0.26	0.32	17.6	0.77	1.14	0.99
13	408-R	Kz-76	Tf	99	47.6	0.058	0.27	0.34	17.5	0.77	0.82	1.02
14	413-R	Kz-31	Al	181	88.9	0.109	0.51	0.60	18.1	0.60	0.99	1.01
15	414-R	Kz-20	St	132	64.0	0.079	0.37	0.45	18.8	0.76	0.99	1.30
16	415-R	Kz-10	St	192	95.6	0.117	0.54	0.64	23.8	0.70	0.89	1.46
17	416-R	Kz-1	St	58.2	27.7	0.034	0.16	0.21	14.8	0.86	0.72	2.77
			av	***128***	**62.5**	**0.077**	**0.36**	**0.43**	***19.0***	***0.74***	***0.91***	***1.32***
			mn	***58.2***	**27.7**	**0.034**	**0.16**	**0.21**	***14.8***	***0.60***	***0.72***	***0.94***
			mx	***192***	**95.6**	**0.117**	**0.54**	**0.64**	***23.8***	***0.86***	***1.14***	***2.77***
						Metamorphic						
18	397-R	Kz-55	Cl-Sl	167	82.2	0.101	0.47	0.56	36.1	0.66	-	-
19	399-R	Kz-21	Cl-Sl	141	68.4	0.084	0.39	0.49	23.6	0.84	0.91	0.88
20	404-R	Kz-61	Cl-Sl	175	86.4	0.106	0.49	0.58	27.6	0.58	0.72	-
21	409-R	Kz-28	Sl	160	77.5	0.095	0.45	0.54	18.4	0.69	1.10	0.53
22	410-R	Kz-69	Cl-Sl	157	76.4	0.094	0.44	0.53	17.7	0.69	0.76	1.46
23	412-R	Kz-66	Cl-Sl	165	80.8	0.099	0.46	0.55	20.3	0.67	1.00	-
			av	***161***	**78.6**	**0.096**	**0.45**	**0.54**	***23.9***	***0.69***	***0.90***	***0.96***
			mn	***141***	**68.4**	**0.084**	**0.39**	**0.49**	***17.7***	***0.58***	***0.72***	***0.53***
			mx	***175***	**86.4**	**0.106**	**0.49**	**0.58**	***36.1***	***0.84***	***1.10***	***1.46***

The Ra-226/U-238 ratios for 10 samples are within ± 10% of the equilibrium value, for 11 samples less than the equilibrium value (0.72-0.89), and in one sample more than the equilibrium value (1.14). The Pb-210/Ra-226 ratio for 8 out of 15 samples was within ± 20% of the equilibrium value, in some samples they differed from the equilibrium value by more than ± 20% - in 6 samples more, and in 1 sample less (Note: the ratios of radionuclide activity were not determined in all samples, so as in a number of samples, the activity of the corresponding radionuclides was less than MDA or was not recorded). The values of radium equivalent activity varied in the range of 58-192 Bq/kg (average value of 136 Bq/kg), while the lowest activity was observed for sedimentary rock samples with an average value of 58.2 Bq/kg, and for samples of magmatic and metamorphic rocks, the average Ra_{eq} values were 103 and 141 Bq/kg, respectively. Similarly, lower absorbed dose rates were observed for sedimentary rock samples (27.7 nGy/h) compared with samples of magmatic (50.0 nGy/h) and metamorphic (68.4 nGy/h) rocks, and, accordingly, the values of the annual effective dose varied from the lowest values for sedimentary rocks (with an average value of 0.034 mSv/y) to high values for magmatic (0.061 mSv/y) and metamorphic (0.084 mSv/y) rocks. It should be noted that the main number of samples (87.0%) in terms of activity in radium equivalent belongs to groups with average radioactivity, and a small number of samples (13.0%) belongs to the group with low radioactivity (Table 3).

As noted above, rock samples were taken in a geological area characterized by a sufficiently complex geotectonic structure. The types of samples collected corresponded to all three main groups of rocks – magmatic, sedimentary and metamorphic. Each of these groups has a specific mineralogical and chemical composition, as well as a mechanism of rock formation, which is mainly associated with a large range of radioactivity concentrations occurring for almost all identified radionuclides, as well as the activity ratios of some radionuclides.

The values of the U-238/U-235 ratio obtained for all samples correspond to the natural value within the margin of error, which, in addition to the methodological aspect, allows us to conclude that there is no pollution by anthropogenic U-235. The ratios of U-238/Th-232 and Ra-226/U-238, which are close to the average and equilibrium values for magmatic samples, allow us to conclude that these primary rocks apparently represent closed systems. To a certain extent, this is confirmed by a slight deviation of the Pb-210/Ra-226 ratio from the equilibrium value (which can be explained by the compacted texture of these rocks).

However, the features of the activity ratio observed in sedimentary and metamorphic samples (which are secondary rocks), in particular, deviations from the average value of the U-238/Th-232 ratio and the equilibrium value of Ra-226/U-238 indicate that these systems were not closed, and in the past, there were various geochemical processes.

For example, in works [20, 21] it is noted that Th isotopes, which in natural environments occur only in a tetravalent form, form compounds practically insoluble in water and are transferred mechanically in the form of stable minerals, while U isotopes occur in both 4- and 6-valence forms - in the 4-valence form, they are close to Th in chemical properties, and in the 6-valence form they are characterized by high chemical activity, and migrate over long distances in the form of solutions with waters. These processes can lead to a decrease in the U-238/Th-232 ratio relative to the average value, especially in metamorphic rocks, where an increased porous structure could contribute to their greater intensification (which was observed, for example, in samples 397-R, 404-R, 409-R, 410-R, and 412-R).

These same factors, as well as the influence of hypergenesis (weathering), can lead to an imbalance in the radioactive series of the family, in particular, between U and Ra. Here, in addition to the above reasons, the influence of differences in the chemical properties of the elements is significant. In particular, the isotope Ra is easily leached and washed away by

waters: in natural formations, Ra-226 often accumulates in quantities exceeding equilibrium with uranium. In the studied samples, deviations from the equilibrium values are relatively small, from which it can be concluded that the above factors played an insignificant role in this region.

The more noticeable deviations of the U-238/Th-232 activity ratios from the average values observed in the work for sedimentary and metamorphic rocks are apparently determined to a greater extent by the processes of hypergenesis. As is known, the Cretaceous period (when most sedimentary rocks were formed) was characterized by the dominance of land on Earth, the climate was hot and dry, deposits of chalk, carbonates, clay shales and others were formed. Mountain systems were formed, and before that, inland seas and vast swamps. All this contributed to the intensification of the process of hypergenesis, intensive leaching of radioactive elements in the process of rock formation or metamorphism. As a result, there was a noticeable decrease in the content of radioactive elements in these rocks. After the Cretaceous period, the climate changed, the temperature dropped, and this factor contributed to a change in the conditions of hypergenesis, which could contribute to an increase in the content of radionuclides during rock formation (which we observe for samples of the Neogene and Quaternary periods).

There is no significant deviation from the equilibrium value in the Pb-210/Ra-226 ratio, which is also noted in [21] for rocks. It can be assumed that the absence of superequilibrium (allochthonous) Pb-210 in rocks (compared to soils, where its noticeable presence is often recorded) is due to their compacted structure, as a result of which, as noted above, radionuclides are in a “sealed” state in them, and radon migration does not take place in them. The slight excess observed in several samples may be associated with the deposition of Pb-210 from atmospheric air.

The insignificant concentrations of the natural radionuclide Be-7 observed in some samples appear in the samples as a result of precipitation, and in some cases can be identified in gamma spectra. Technogenic radionuclide Cs-137 also gets onto samples as a result of atmospheric precipitation. Usually, the presence of Cs-137 in natural objects (for example, in soil) is associated, as mentioned above, with the Chernobyl disaster. Over the past period, its number has decreased as a result of both disintegration and migration. In the conducted studies, it was observed in several samples in insignificant concentrations (compared with soils where its concentration is noticeably higher), which is apparently due to the intensive process of leaching from the surface of the samples.

The obtained activity values in radium equivalent are noticeably lower than the recommended value of 370 Bq/kg [2, 3].

CONCLUSION

In this work, the radionuclide content and activity concentration of gamma-emitting radionuclides were studied in various rock samples collected on the territory of the Kazbegi municipality.

As a result of the conducted studies, it was found that up to 22 radionuclides are mainly detected in the selected rock samples, in particular: family Th-232 - Ac-228, Th-228, Ra-224, Pb-212, Bi-212, Tl-208 (a total of 6 radionuclides); family U-238 – Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, Pb-210 (7 radionuclides in total); family U-235 - U-235, Th-231, Th-227, Ra-223, Rn-219, Pb-211 (6 radionuclides in total); other natural radionuclides Be-7, K-40, technogenic radionuclide Cs-137. For the studied samples, the average activity values of the radionuclides of the Th-232 and U-238 families are approximately the same (42.4 Bq/kg and 31.1 Bq/kg, respectively) and significantly exceed the average activity of the radionuclides of

the U-235 family (1.91 Bq/kg). The activity of radionuclide K-40 varies in the range of 204-1069 Bq/kg (the average value was 632 Bq/kg); in some samples, a small amount of natural radionuclide Be-7 and technogenic radionuclide Cs-137 was recorded. The main number of samples (87.0%) in terms of activity level in radium equivalent belongs to the groups with medium radioactivity, and a small number of samples (13.0%) belongs to the group with low radioactivity. The obtained activity values in radium equivalent are noticeably lower than the recommended value of 370 Bq/kg.

Thus, the radioecological situation of the territory of the Kazbegi municipality in terms of radioactivity of rocks can be assessed as normal, not posing a danger to the resident population.

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