

Radionuclides in Samples of Soil of Different Types in the Kvemo Kartli Region (Georgia)

Nodar Kekelidze^{1, 2, 3}, Teimuraz Jakhutashvili¹, Bezhan Tutberidze⁴, Eremia Tulashvili^{1, *}, Mariam Akhalkatsishvili⁴, Lela Mtsariashvili¹

¹Material Research Institute, Faculty of Exact and Natural Sciences, Ivane Javakhishvili Tbilisi State University, Tbilisi, Georgia

²Semiconductor Materials Science Laboratory, Ferdinand Tavadze Metallurgy and Materials Science Institute, Tbilisi, Georgia

³Department of Engineering Physics, Faculty of Informatics and Control Systems, Georgian Technical University, Tbilisi, Georgia

⁴Department of Geology, Faculty of Exact and Natural Sciences, Ivane Javakhishvili Tbilisi State University, Tbilisi, Georgia

Email address

eremia.tulashvili@tsu.ge (E. Tulashvili)

*Corresponding author

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Abstract: The aim of the present work is to determine the radionuclide content and radioactivity in various types of soil in the territory of Kvemo Kartli (Georgia). Within this territory samples of soil of various types [cinnamonic (Cn), cinnamonic leached (Cn-Ld), cinnamonic calcareous (Cn-Cr), grey cinnamonic dark (GCD), chernozems (Cz), and brown forest weakly unsaturated (BFWU)] were selected at 28 locations. Up to 22 radionuclides were identified in these samples. The mean (range) concentration of radionuclides of the Th-232 family (six identified radionuclides in total) was 27.0 (12.9-64.9) Bq/kg, that of the U-238 family (seven radionuclides in total) was 24.0 (10.2-47.9) Bq/kg, and that of the U-235 family (six radionuclides in total) was 1.1 (0.49-2.3) Bq/kg. Individual radionuclides were also identified: Be-7 [28.9 (16.1-42.6) Bq/kg], K-40 [475 (290-942) Bq/kg], and Cs-137 [15.6 (0.2-88.1) Bq/kg]. Mean radium equivalent activity was 102 (57.1-210) Bq/kg; therein, the greatest values were observed for soil GCD, Cn-Ld, and Cn and slightly smaller ones for Cz, Cn-Cr and BFWU. Mean annual effective dose was 0.058 (0.035-0.122) mSv/y. Some features of radionuclides distribution were marked, in particular, depending on the soil type and sampling location. Some activity ratios of radionuclides were considered, in particular, U-238/U-235, U-238/Th-232, Ra-226/U-238, and Pb-210/Ra-226 (in the last case appreciable deviations in the positive direction from the equilibrium value were observed – up to 6.87 with an average value of 2.29; therein, a certain correlation with features of geotectonic structure in the studied territory was marked). Comparison with reference data was carried out.

Keywords: Radionuclides, Soil, Activity Concentration, Activity Ratio, Exposure Dose, Georgia

1. Introduction

Natural and technogenic radioactivity of soil is one of the main components of the Earth's radioactive background. As numerous studies have shown, the radioactivity in various areas of the globe differs considerably – from units up to hundreds of Bq/kg, exposing the population to constant radiation levels that can cause various illnesses. In many works the activity ratios of various radionuclides have been investigated, indicating a certain level of interest in understanding various geochemical processes in rocks and soils. For example, the activity ratio, U-238/U-235, which is

a constant equal to 21.7 in rocks and soils [1, 2] is often investigated for the purpose of identification of technogenic pollution by uranium. A number of other activity ratios, for example, U-238/Th-232, and also Ra-226/U-238, Pb-210/Ra-226 [reflecting disbalance in radioactive chain U-238 in zones of hypergenesis (weathering)], are often a subject of investigation [3-5]. Therefore, it is important that such studies be carried out in each territorial region.

For example, Charro and Moyano [6] studied soil samples selected in the territory close to the small village of Villar de Peralonso in central-western Spain (Salamanca province). The activities of five natural radionuclides (U-238, Ra-226, Pb-210, Th-232, and Ra-224) were measured in 13 top soils. The

authors noted that investigated soils are rich in Ra-226 and Pb-210, given the concentration of its isotope of origin, U-238. In order to perform a better analysis of this feature, the ratios of activity Ra-226/Pb-210, U-238/Ra-226, U-238/Pb-210, and U-238U/Th-232 in the soils were evaluated for all the samples.

The aim of another study [7] was to investigate the specific activity of radionuclides in soils of the subdivisions Murree and Kotli Sattian (Himalayan Mountain range) Pakistan; the activity concentrations of primordial radionuclides Ra-226, Th-232, and K-40 as well as technogenic radionuclide Cs-137 were measured in soil samples; the Th-232/Ra-226 activity ratio (i.e., progeny pair Ac-228/Pb-214) was calculated to assess the maintenance of the proportionality within the Th-232 and Ra-226 decay series, which in most environmental samples is approximately 1.1. Values of radium equivalent activity were calculated and varied from 107.3 to 147.8 Bq/kg, with an average value of 126.6 Bq/kg, which is 66% lower than safe limit of 370 Bq/kg [8, 9].

In one work [10], depth distributions studies of radionuclides Th-232, U-238, Ra-226, K-40, Pb-210, and Cs-137 were carried out in the moraine and the colluvium soils selected in the inner Nordfjord region located in western Norway on the western side of the Jostedalsgreen ice cap.

In another work [11] soil samples collected from undisturbed sites (of an oil field) in the Niger Delta region of Nigeria were studied. The specific gamma activity concentration of Ra-226, Th-232 and K-40 was determined, and values of radium equivalents were calculated. The authors noted that the linear fit of all the measured parameters was significantly different from unity (1), which shows that the concentration of NORM were mainly influenced by oil exploration and production activities and not by the geological constituent of the area.

Activity concentrations of radionuclides U-238, Th-232, Ra-226, K-40, Cs-137, Be-7, and U-235 in soil of the city of Yalova, northwestern Turkey, were determined by Kapdan *et al.* [12]; several radiological parameters related to terrestrial gamma-radioactivity, including the annual effective dose equivalent, were calculated.

In Georgia regular studies of natural (and technogenic) radioactivity have not been carried out. Rather detailed studies of radioactivity in various environmental materials were carried out in 1986 after the failure of the Chernobyl atomic power station and were mainly concerned with technogenic radionuclides [13-15]. In these works, it was shown that during this period in the territory of western Georgia, essentially in the strip adjoining the sea, high concentrations of technogenic radionuclides were observed (in particular, Cs-137 concentration was detected from several hundreds to some thousands of Bq/kg). In other notable works [16, 17] the results of research on radiation condition in coastal water areas of the Black Sea during a later period are reported; in particular, the presence of seven natural (Ac-228, Ra-226, Bi-214, Pb-214, Pb-212, Pb-210, and K-40) and one technogenic radionuclide (Cs-137) has been found in soil in some areas of Achara (Batumi, Gonio, Sarpi, Chakvi, and Kvartli). Some results of the last period are given in a study by Kekelidze *et al.* [18].

In the present work we report the results of radioactivity research on different soil types within the territory of Kvemo Kartli region, located in the southeastern part of Georgia.

2. Materials and Methods

2.1. Study Area

The studied territory is located to the southwest from city of Tbilisi, the main city of Georgia. Mountainous terrain is the prevailing landform of the territory. A wide variety of soil types is observed: different types of cinnamonic soils, chernozem, etc. The region is characterized by sufficiently complex geotectonic structure; in particular, it is located in the folded (folded-thrust) system of Lesser Caucasus (III) – the so-called structural-morphological unit of the first order. The studied area, in turn, is divided into some tectonic units of the second, third and fourth orders, in particular, the following:

- a. The southern subzone (III_1^4) is located in the Achara-Trialeti folded zone (III_1), a mountainous area (600 – 1000 m above sea level), elongated in the latitudinal direction between the southeast coast of the Black Sea and the valley of the river Iori structures of various morphology.
- b. The Bolnisi subzone (of weakly folded sedimentary cover, III_2^2) is located to the south of the southern subzone, in the Artvin-Bolnisi zone (block mass, III_2). It includes Khrami crystalline massif with volcanic-sedimentary formations and the territory located to the south, up to the Loki massif with thick volcanic formations. Four large structures of the fourth order – so-called blocks – are distinguished in the Bolnisi subzone: 1 – Khrami (bulge of pre-Jurassic formation), 2 – Tetrtskaro-Asureti, 3 – Madneuli-Poladauri, and 4 – Marneuli [19].

The region is characterized by the developed underground waters, in particular, located in subzone III_1^4 is the so-called Tbilisi water pressure system of fissure and fissure-karstic waters. Here, underground interstitial waters and underground waters of deep circulation are observed, which are mainly developed in volcanic sedimentary rock strata of the Middle Eocene and carbonate rock strata of the Upper Cretaceous. Located in the western part of subzone III_2^2 is the so-called area of underground fissure waters of Akhaltsikhe lava formations, which is characterized by high abundance of water in many places, and in the eastern part, there is the so-called area of underground fissure waters of the east slope of the Javakheti ridge where water-bearing complexes of crystal rocks of various formations are distinguished [20].

In this territory there are 28 sampling locations nearby settlements:

- a. in subzone III_1^4 – village Pantiani (Ph), village Didgori (Dg), township Manglisi (Ms), village Tskhrakudaani (Td), village Algeti (Al), and village Gokhnari (Gh);
- b. in subzone III_2^2 :
 1. in the northern part: villages Kumisi (Kf), Koda (Kd), Jorjiashvili (Jr), Menkalisi (Mk), Didi Kldeisi (Kz), Trialeti (Tb), city Tsalka (Tl), and village Imera (Im),
 2. in the southern part: city Marneuli (Mn), villages

2.2. Sampling and Analysis

2.2.1. Sampling

Samples were collected using a special hand auger directly into plastic containers. After drying under laboratory conditions, samples were ground and sieved to ensure homogenization. Samples weights were measured and their specific weights were determined. Then the samples were dried at 105-110 °C to a constant weight, and their bulk density was determined. These values were used to describe sample geometry. The samples were sealed in a Marinelli beaker (volume 1.0 L) and stored for more than 4 weeks to achieve secular equilibrium between Ra-226 and Rn-222.

2.2.2. Gamma-Radiation Activity Measurement

Measurements were carried out using a Canberra GC2020 gamma spectrometer with a semi-conductor germanium detector with a relative efficiency of 24%. The gamma spectra acquisition time was 72 h. For the analysis, the software Genie-2000 S500 was used with additional modules, in particular, S506 – the Interactive Fit Program. The activity concentrations of Th-232 were determined (averaged values were reported for Ac-228, Ra-224, Pb-212, Bi-212, and Tl-208, of which the determination error varied from 2.5% to 4.1%), U-238 (by the Th-232 line of 63.3 keV with an error in the diapason varying from 7.6% to 18.1%), Ra-226 and U-235 (by the 186 keV line, which was divided using the S506 program with error of 11.3%-25.4% for Ra-226, and 6.5%-10.3% for U-235), Pb-214 and Bi-214 (with error from 2.3% to 5.9%), Pb-210 (by the 46.54 keV line with an error from 9.5% to 18.7%). Also identified were Be-7, K-40, and the technogenic radionuclide Cs-137. In samples “super-equilibrium” (allochthonous) Pb-210 (Pb_{al}) was often observed, the value of which was determined as the difference between measured activity values of Pb-210 and Ra-226 [3]. Considering, that their values (ratios), as it was reported in a number of works [4, 21, 22], can be related to the geotectonic features of investigated territory, the generalized parameters (average (av), minimal (mn), and maximal (mx) values of activity concentration of Ra-226 and Pb-210, activity ratios) by various geotectonic zones and subzones were also analyzed.

With an account taken of the influence of matrix composition, the chemical composition of samples was determined on the basis of reference data [23], which were then used with the special software (LabSOCS) for efficiency calibration at the calculated activity concentration. The LabSOCS system allows the creation of calibrations by laboratory quality efficiency without application of radioactive calibration sources. For radionuclides identification, a special library containing lines of 41 radionuclides and other specific sources (in total 351 lines) was used. The database NuDat [24] was used for library creation. For the activity calculation, background radiation was subtracted.

Assessment of radium equivalent activity Ra_{eq} (Bq/kg) and annual effective dose equivalent AEDE (mSv/y) depending

on the soil type was carried out under formulas [25]:

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

where A_U , A_{Th} , and A_K are the activity concentrations (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 in 1 y. (=8760 h), k_1 – the factor to convert effective dose rate into the absorbed dose rate in the air for adults, 0.7×10^3 mSv/Gy, k_2 – outdoor occupancy factor (the fraction of time spent in the open air) which equals to 0.2, D – absorbed dose rate D (nGy/h):

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

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

When allochthonous Pb was present, the contribution of its “allochthonous” parents Pb-214 and Bi-214 (and, accordingly, the activity Ra_{eq-al} of allochthonous Ra-226) to radioactivity was considered (under the assumption that its concentration is connected with excess radon in a soil layer), the contribution of which, according to Saito and Jacob [26], makes up a part equal to 98.5% of the gamma flow of the energy of U-238 radionuclides. In this case the term equal to $0.985 \cdot A_{Ra-al}$ was added into the calculation formula (1) where A_{Ra-al} is equal to $A_{Pb-al} = A_{Pb} - A_{Ra}$ [3]. The similar term ($0.456 \cdot A_{Ra-al}$) was added into equation (3) for the absorbed dose rate D_{al} and, accordingly, was considered for calculation of $AEDE_{al}$.

For samples characterization by degree of radioactivity taking into account the accepted limiting value $Ra_{eq} - 370$ Bq/kg (equivalent to γ -radiations dose of 1.5 mSv/y) [27] there were established several groups of samples by value of radium equivalent activity, in particular: 1st group - not radioactive samples with activity no more than 30 Bq/kg; 2nd group – with a low radioactivity in the range from 30 to 100 Bq/kg; 3rd group – with an average radioactivity in the range from 100 to 300 Bq/kg; 4th group – samples with the raised radioactivity¹ in the range from 300 to 1000 Bq/kg. The technique is described in more detail in works [28, 29].

3. Results

Based on the results of the gamma spectral analysis, up to 22 radionuclides were identified in samples: the Th-232 family (Ac-228, Th-228, Ra-224, Pb-212, Bi-212, and Tl-208), the U-238 family (Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, and Pb-210), the U-235 family (U-235, Th-231, Th-227, Ra-223, Rn-219, and Pb-211), the individual radionuclides Be-7, K-40, and the techogenic radionuclide Cs-137 (some specific gamma lines were also identified, originated as a result of cosmic rays interacting with the

¹ Such samples were not observed in the present work.

material of the detector or the sample).

The activity of identified radionuclides of the different families varied from 0.49 Bq/kg for U-235 to 64.9 Bq/kg for Th-232. Among individual radionuclides K-40 had the greatest activity (up to 942 Bq/kg). The activity of several radionuclides (namely, those of the U-235 family) in some samples was below the minimal detectable activity (MDA).

Activity concentrations of the main radionuclides in the studied samples, equivalent radioactivity with no account taken of allochthonous Pb-210_{al} (Ra_{eq}) and taking this into account (Ra_{eq-al}) and, accordingly, annual effective dose (AEDE and AEDE_{al}), activity ratios, their average (av), minimal (mn) and maximal (mx) values, and other data are given in Tables 2-6.

Table 2. Activity concentrations (Bq/kg) of families' radionuclides Th-232, U-238, Ra-226, Pb-210, and U-235, radionuclides Be-7, K-40, and Cs-137, equivalent activities with no account taken of allochthonous Pb-210_{al} (Ra_{eq}) and taking this into account (Ra_{eq-al}), annual effective doses (AEDE and AEDE_{al}), activity ratios U-238/U-235, U-238/U-235, Ra-226/U-238, and Pb-210/Ra-226.

#	Tct	L	SN	ST	Th-232	U-238	Ra-226	Pb-214	Bi-214	Pb-210	U-235	Be-7	K-40	Cs-137
1	III ₁ ⁴	Ph-1	163	Cn-Ld	20.7	20.5	26.7	23.6	20.9	64.3	1.0	42.6	433	24.8
2	-“-	Dg-1	165	Cn-Ld	20.6	28.1	25.7	24.5	23.4	50.5	1.3	—	417	5.7
3	-“-	Tp-1	167	Cn-Ld	12.9	16.6	15.2	14.9	13.5	26.1	0.75	<M	333	4.1
4	-“-	Ms-1	168	Cn-Ld	22.2	34.2	40.7	36.5	34.9	51.0	1.63	<M	535	5.1
5	-“-	Td-2	178	Cn	17.9	24.0	26.0	25.1	24.5	28.3	1.12	27.9	484	2.0
6	-“-	Al-1	172	Cn	18.7	18.5	25.3	22.0	20.9	55.5	0.87	32.6	452	10.3
7	-“-	Al-5	176	Cn	26.6	29.8	28.4	26.5	26.0	45.1	1.38	—	668	1.1
8	-“-	Gh-2	179	Cz	22.3	27.9	25.9	29.7	28.4	61.2	1.4	—	387	14.0
9	III ₂ ²	Kf-1	64	GCD	21.2	47.9	41.2	43.6	43.5	39.5	2.3	—	382	4.1
10	-“-	Kd-1	63	GCD	21.2	22.8	29.1	23.1	22.1	51.6	1.03	—	468	9.5
11	-“-	Jr-1	201	Cn-Cr	20.7	21.1	37.9	33.5	31.6	61.3	0.99	<M	456	17.4
12	-“-	Mk-1	200	BFWU	27.2	17.3	26.7	22.2	21.0	86.1	0.77	25.2	439	24.6
13	-“-	Kz-5	198	Cn-Ld	64.9	44.9	51.0	50.0	48.6	104	2.08	—	942	14.9
14	-“-	Kz-4	196	Cn-Ld	26.0	16.4	22.9	19.4	19.1	132	0.74	<M	360	6.2
15	-“-	Tb-3	192	BFWU	19.2	10.2	13.7	11.7	11.2	49.9	0.49	16.1	290	0.2
16	-“-	Tb-1	190	Cz	43.8	25.0	43.2	38.3	37.6	95.1	1.11	<M	473	30.5
17	-“-	Tl-2	189	Cz	26.8	20.1	25.8	23.5	21.2	177	0.94	—	440	57.5
18	-“-	Im-2	187	Cz	29.2	17.3	26.4	24.6	23.8	46.8	0.83	—	374	5.3
19	-“-	Im-1	185	Cz	33.9	21.3	31.5	31.1	29.4	57.6	0.98	—	459	9.2
20	-“-	Gh-1	184	Cz	29.5	17.1	22.7	20.8	20.4	19.5	0.78	—	375	0.7
21	-“-	Mn-1	62	GCD	29.0	20.6	41.1	38.7	37.6	64.5	0.92	—	502	25.4
22	-“-	Tm-4	61	GCD	27.4	27.6	27.8	23.3	22.9	64.5	1.21	—	469	15.5
23	-“-	Kc-2	55	Cn-Cr	23.0	21.6	23.1	23.5	22.6	137	1.05	—	670	88.1
24	-“-	Sd-2	53	Cn-Cr	19.8	21.1	27.0	22.8	21.4	38.3	0.96	<M	406	4.0
25	-“-	Pn-2	51	Cn	39.9	42.2	38.8	38.6	37.1	27.3	1.98	<M	752	1.0
26	-“-	Gn-2	47	Cn	37.8	20.0	41.2	36.0	34.2	43.1	0.97	—	519	14.1
27	-“-	Dm-9	45	Cn-Ld	22.4	18.0	19.1	20.3	19.8	30.7	0.86	—	434	10.0
28	-“-	Dm-4	41	Cn-Ld	30.8	20.2	29.7	26.8	25.6	74.0	0.96	<M	376	30.9
av					27.0	24.0	29.8	27.7	26.5	63.6	1.1	28.9	475	15.6
mn					12.9	10.2	13.7	11.7	11.2	19.5	0.49	16.1	290	0.2
mx					64.9	47.9	51.0	50.0	48.6	177	2.3	42.6	942	88.1

Table 2. Continued.

#	Tct	L	SN	ST	Pb-210 _{al}	Ra_{eq}	Ra_{eq-al}	U-238 /U-235	U-238 /Th-232	Ra-226 /U-238	Pb-210 /Ra-226	AEDE mSv/y	AEDE _{al} mSv/y
1	III ₁ ⁴	Ph-1	163	Cn-Ld	37.6	86.7	124	21.4	0.99	1.30	2.41	0.049	0.070
2	-“-	Dg-1	165	Cn-Ld	25	84.3	109	21.5	1.36	0.91	1.97	0.053	0.067
3	-“-	Tp-1	167	Cn-Ld	11	57.1	68	22.2	1.28	0.92	1.71	0.036	0.042
4	-“-	Ms-1	168	Cn-Ld	10	110	120	21.0	1.54	1.19	1.25	0.063	0.069
5	-“-	Td-2	178	Cn	2	85.5	88	21.4	1.34	1.08	1.09	0.052	0.053
6	-“-	Al-1	172	Cn	30	83.6	113	21.3	0.99	1.37	2.20	0.048	0.065
7	-“-	Al-5	176	Cn	17	113	130	21.6	1.12	0.95	1.59	0.071	0.080
8	-“-	Gh-2	179	Cz	35	84.8	120	20.2	1.25	0.93	2.37	0.052	0.072
9	III ₂ ²	Kf-1	64	GCD	-2	98.3	97	20.8	2.26	0.86	0.96	0.063	0.062
10	-“-	Kd-1	63	GCD	23	92.2	114	22.2	1.08	1.28	1.77	0.053	0.065
11	-“-	Jr-1	201	Cn-Cr	23	100	123	21.3	1.02	1.80	1.62	0.051	0.064
12	-“-	Mk-1	200	BFWU	59	96.4	155	22.5	0.64	1.54	3.22	0.053	0.086
13	-“-	Kz-5	198	Cn-Ld	53	210	262	21.6	0.69	1.14	2.04	0.122	0.152
14	-“-	Kz-4	196	Cn-Ld	109	85.3	192	22.0	0.63	1.40	5.75	0.047	0.108
15	-“-	Tb-3	192	BFWU	36	61.4	97	20.7	0.53	1.34	3.65	0.035	0.055

#	Tct	L	SN	ST	Pb-210 _{al}	Ra _{eq}	Ra _{eq-al}	U-238 /U-235	U-238 /Th-232	Ra-226 /U-238	Pb-210 /Ra-226	AEDE mSv/y	AEDE _{al} mSv/y
16	-“-	Tb-1	190	Cz	52	139	190	22.5	0.57	1.73	2.20	0.071	0.100
17	-“-	Tl-2	189	Cz	152	95.0	244	21.4	0.75	1.28	6.87	0.054	0.139
18	-“-	Im-2	187	Cz	20	94.4	114	20.9	0.59	1.53	1.77	0.051	0.062
19	-“-	Im-1	185	Cz	26	112	138	21.7	0.63	1.48	1.83	0.061	0.075
20	-“-	Gh-1	184	Cz	-3	91.1	88	22.0	0.58	1.32	0.86	0.051	0.049
21	-“-	Mn-1	62	GCD	23	118	141	22.4	0.71	1.99	1.57	0.059	0.072
22	-“-	Tm-4	61	GCD	37	99.7	136	22.8	1.01	1.00	2.32	0.060	0.081
23	-“-	Kc-2	55	Cn-Cr	114	103	216	20.5	0.94	1.07	5.96	0.064	0.128
24	-“-	Sd-2	53	Cn-Cr	11	83.7	95	21.9	1.07	1.28	1.42	0.047	0.054
25	-“-	Pn-2	51	Cn	-12	148	137	21.3	1.06	0.92	0.70	0.092	0.086
26	-“-	Gn-2	47	Cn	2	132	133	20.6	0.53	2.05	1.05	0.066	0.067
27	-“-	Dm-9	45	Cn-Ld	12	81.5	93	21.0	0.80	1.06	1.61	0.049	0.056
28	-“-	Dm-4	41	Cn-Ld	44	100	144	21.1	0.66	1.47	2.49	0.054	0.078
				av	33.9	102	135	21.5	0.95	1.29	2.29	0.058	0.077
				mn	-11.6	57.1	67.8	20.2	0.53	0.86	0.70	0.035	0.042
				mx	152	210	262	22.8	2.26	2.05	6.87	0.122	0.152

Table 3. Generalized data – average (av), minimal (mn), and maximal (mx) values of concentrations of radionuclides of families (Th-232, U-238 and Ra-226, U-235), and individual radionuclides (Be-7, K-40, and Cs-137) – depending on soil type (ST).

#	ST (SN)	Th-232			U-238			Ra-226			Pb-210		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	GCD (64, 63, 62, 61)	24.7	21.2	29.0	29.7	20.6	47.9	34.8	27.8	41.2	55.0	39.5	64.5
2	Cn-Ld (163, 165, 167, 168, 198, 196, 45, 41)	50.5	12.9	64.9	24.9	16.4	44.9	28.9	15.2	51.0	66.5	26.1	132
3	Cn (178, 172, 176, 51, 47)	28.2	17.9	39.9	26.9	18.5	42.2	31.9	25.3	41.2	39.8	27.3	55.5
4	Cz (179, 184, 185, 187, 189, 190)	26.5	22.3	43.8	18.4	17.1	27.9	25.1	22.7	43.2	65.4	19.5	177
5	Cn-Cr (201, 55, 53)	21.2	19.8	23.0	21.3	21.1	21.6	29.3	23.1	37.9	79.0	38.3	137
6	BFWU (200, 192)	24.7	21.2	29.0	29.7	20.6	47.9	34.8	27.8	41.2	55.0	39.5	64.5

Table 3. Continued.

#	ST (SN)	U-235			Be-7			K-40			Cs-137		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	GCD (64, 63, 62, 61)	1.4	0.92	2.3	—	—	—	455	382	502	13.6	4.1	25.4
2	Cn-Ld (163, 165, 167, 168, 198, 196, 45, 41)	1.2	0.74	2.1	42.6	42.6	42.6	479	333	942	12.7	4.1	30.9
3	Cn (178, 172, 176, 51, 47)	1.3	0.87	2.0	30.2	27.9	32.6	575	452	752	5.7	1.0	14.1
4	Cz (179, 184, 185, 187, 189, 190)	0.86	0.78	1.4	—	—	—	358	374	473	16.7	0.7	57.5
5	Cn-Cr (201, 55, 53)	1.0	0.96	1.1	—	—	—	511	406	670	36.5	4.0	88.1
6	BFWU (200, 192)	1.4	0.92	2.3	—	—	—	455	382	502	13.6	4.1	25.4

Table 4. Generalized data – average (av), minimal (mn), and maximal (mx) values of equivalent activities with no account taken of allochthonous Pb-210_{al} (Ra_{eq}) and taking this into account (Ra_{eq-al}), activity ratios, and annual effective doses (AEDE and AEDE_{al}) – depending on soil type (ST).

#	ST (SN)	Ra _{eq}			Ra _{eq-al}			U-238/Th-232			Ra-226/U-238		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	GCD (64, 63, 62, 61)	102	92.2	118	122	96.6	141	1.26	0.71	2.26	1.28	0.86	1.99
2	Cn-Ld (163, 165, 167, 168, 198, 196, 45, 41)	102	57.1	210	139	67.8	262	0.99	0.63	1.54	1.17	0.91	1.47
3	Cn (178, 172, 176, 51, 47)	112	83.6	148	120	87.8	137	1.01	0.53	1.34	1.28	0.92	2.05
4	Cz (179, 184, 185, 187, 189, 190)	88.0	84.8	139	128	88.0	244	0.62	0.57	1.25	1.18	0.93	1.73
5	Cn-Cr (201, 55, 53)	95.4	83.7	103	144	94.8	216	1.01	0.94	1.07	1.38	1.07	1.80
6	BFWU (200, 192)	78.9	61.4	96.4	126	97.0	155	0.59	0.53	0.64	1.44	1.34	1.54

Table 4. Continued.

#	ST (SN)	Pb-210/Ra-226			AEDE, mSv/y			AEDE _{al} mSv/y		
		av	mn	mx	av	mn	mx	av	mn	mx
1	GCD (64, 63, 62, 61)	1.66	0.96	2.32	0.059	0.053	0.063	0.070	0.062	0.081
2	Cn-Ld (163, 165, 167, 168, 198, 196, 45, 41)	2.40	1.25	5.75	0.059	0.036	0.122	0.080	0.042	0.152
3	Cn (178, 172, 176, 51, 47)	1.32	0.70	2.20	0.066	0.048	0.092	0.070	0.053	0.086
4	Cz (179, 184, 185, 187, 189, 190)	2.27	0.86	6.87	0.049	0.051	0.071	0.071	0.049	0.139
5	Cn-Cr (201, 55, 53)	3.00	1.42	5.96	0.054	0.047	0.064	0.082	0.054	0.128
6	BFWU (200, 192)	3.43	3.22	3.65	0.044	0.035	0.053	0.071	0.055	0.086

Table 5. Generalized data – average (av), minimal (mn), and maximal (mx) values of activity concentrations (Bq/kg) of U-238, Ra-226, and Pb-210, equivalent activity (Ra_{eq} , Bq/kg), activity ratios – depending on tectonic structure (Tct).

#	Tct	U-238			Ra-226			Pb-210			Ra_{eq}		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	III ₁ ⁴	24.9	16.6	34.2	26.7	15.2	40.7	47.7	26.1	64.3	88.1	57.1	113
2	III ₂ ²	23.6	10.2	47.9	31.0	13.7	51.0	70.0	19.5	177	107	61.4	210
3	2	30.6	21.1	47.9	36.1	29.1	41.2	50.8	39.5	61.3	96.6	92.2	99.5
4	1	21.1	10.2	44.9	29.3	13.7	51.0	85.3	19.5	177	109	61.4	210
5	4	24.1	20.6	27.6	34.4	27.8	41.1	64.5	64.5	64.5	109	99.7	118
6	3	26.2	20.0	42.2	32.5	23.1	41.2	61.5	27.3	137	117	83.7	148

Table 5. Continued.

#	Tct	U-238/Th-232			Ra-226/U-238			Pb-210/Ra-226		
		av	mn	mx	av	mn	mx	av	mn	mx
1	III ₁ ⁴	1.23	0.99	1.54	1.08	0.91	1.37	1.82	1.09	2.41
2	III ₂ ²	0.84	0.53	2.26	1.38	0.86	2.05	2.48	0.70	6.87
3	2	1.45	1.02	2.26	1.31	0.86	1.80	1.45	0.96	1.77
4	1	0.62	0.53	0.75	1.42	1.14	1.73	3.13	0.86	6.87
5	4	0.86	0.71	1.01	1.50	1.00	1.99	1.95	1.57	2.32
6	3	0.90	0.53	1.07	1.33	0.92	2.05	2.28	0.70	5.96

Table 6. Distribution of average value Ra_{eq-av} (Bq/kg) of equivalent activity Ra_{eq} by the activity group (GA), their quantity (N_s) and percentage (r, %).

#	GA	Ra_{eq} (Bq/kg)	Ra_{eq-av} (Bq/kg)	N_s	r (%)
1	II	30-100	88.0	18	64.3
2	III	100-300	128	10	35.7

3.1. General Characteristics

Within families of radionuclides, activity varied within sufficiently wide limits (Table 2). In particular, the range (mean) was 12.9-64.9 (27.0) Bq/kg for Th-232, 10.2-47.9 (24.0) Bq/kg for U-238, and 0.49-2.3 (1.1) Bq/kg for U-235. The activity of K-40 varied from 290 to 942 Bq/kg (mean, 475). Be-7 was measured in five samples and ranged from 16.1 to 42.6 Bq/kg. Cs-137 was measured in all samples and ranged from 0.2 to 88.2 Bq/kg (mean, 15.6). The activity ratio U-238/U-235 (within $\pm 10\%$) was 21.7 (accepted for natural objects). The ratios U-238/Th-232 deviated (more than 10%) from the average value of 0.81 (for closed systems) in the positive direction (up to 2.26 and 2.05, accordingly) as well as towards decrease (up to 0.53 and 0.86); ratios Ra-226/U-238 almost in all types of soil correspond (within $\pm 10\%$) or exceed (up to 2.05) equilibrium value (1.00). The ratio Pb-210/Ra-226 in almost all samples was appreciably (more than 20%²) greater than the equilibrium value (the greatest value of 6.87). Radionuclides in chain Th-232 - Tl-208 were essentially in equilibrium (except for Th-228, for which the determination error was appreciably more than for other radionuclides). The greatest proportion of samples (64.3%), by the level of equivalent activity, belonged to the group with low radioactivity, and a small proportion of samples (35.7%) belonged to the group with average values of equivalent activity (Table 6). It is worthy of note that if one considers allochthonous activity, the majority of samples (75.0%) were in the average group.

3.2. Dependence on the Type

The highest values of equivalent activity (Table 3, Table 4) were observed for soil types GCD, Cn-Ld, and Cn, with average values from 102 to 112 Bq/kg and rather less for soils Cz, Cn-Cr, and BFWU, from 78.9 to 95.4 Bq/kg [note: for soil type Cn-Ld, sample 198 (selected at the location Kz-5) was notable for having an equivalent activity of 210 Bq/kg, an appreciably greater activity than all other samples].

The activity ratio U-238/Th-232 for soils GCD essentially corresponded (within $\pm 10\%$) or exceeded (up to 2.26) the average value of 0.81. For soils Ch-Ld, Cn, and Cz values less than average value as well as greater values were observed, whereas values in soils Cn-Cr were greater (up to 1.07) and in soils BFWU were less (up to 0.53) than the average value. The ratio Ra-226/U-238 in almost all soil types met (within $\pm 10\%$) or exceeded the equilibrium value (note: a similar analysis for ratio Pb-210/Ra-226 was not carried out).

3.3. Dependence on the Tectonic Zone

The average value of Ra-226 activity and its equivalent activity in subzone III₁⁴ (Table 5) was rather low (26.7 and 88.1 Bq/kg, respectively) in comparison with subzone III₂² (31.0 and 107 Bq/kg, respectively). This difference was very noticeable for Pb-210 and ratio Pb-210/Ra-226: 47.7 and 70.0 Bq/kg and 1.82 and 2.48, respectively. It was of particular interest that the greatest values of the ratio Pb-210/Ra-226 were observed for samples 55, 189, and 196 (and also 200, 192, and 190), from 5.75 to 6.87 (2.20-3.65), selected at locations Kc-2, Tl-2, and Kz-4, (Mk-1, Tb-3, and Tb-1) located in the northern part of subzone III₂² (Figure 1,

2 The range of limits is expanded, because determination error of Pb-210 reached up to 20%.

Table 2) nearby corresponding tectonic borders between subzones and units. The ratio U-238/Th-232 for samples from subzone III₁⁴ exceeded the average value (0.81), while these values for subzone III₂² were both less and greater than the average value. The ratio Ra-226/U-238 for subzone III₁⁴ on the average corresponded to the equilibrium value, and for subzone III₂² the higher values were dominant (up to 2.05).

Activity distribution depending on the blocks in the zone III₂² can be of the certain interest. Evidently (see Table 5), the greatest equivalent activity (117 Bq/kg) was observed in the block 3, the least activity (96.6 Bq/kg) was observed in the block 2.

3.4. Radiological Parameters

The minimal and maximal values of annual effective dose differed by more than 3-fold and were in the range 0.035-0.122 mSv/y (Table 2). These values (as well as equivalent activity) increased considerably (annual effective dose in the range 0.042-0.152 mSv/y) under the assumption that allochthonous Pb-210 is caused by excess soil radon.

4. Discussion

All identified radionuclides, except for Cs-137, were naturally occurring. They were also observed in soil in some regions of western Georgia [30].

The content and concentration of naturally occurring radionuclides, in general, corresponded to those usually observed [4] for various soils.

The observed peculiarities of the activity ratios, in particular, the dominance of U-238/Th-232 ratio relative to the average value, may be related to various geochemical processes U and Th under identical environmental conditions. For example, Kogan *et al.* [4] and Titaeva [5] noted that Th isotopes in nature occur only in the tetravalent form; their compounds are practically insoluble in water, and they are transferred mechanically in the form of stable minerals, whereas U isotopes occur in the tetravalent form as well as in the hexavalent form. In the tetravalent form their chemical properties are similar to those of Th, but in the hexavalent form they have greater chemical activity and can migrate long distances in the form of water solutions. As noted above, the investigated region is characterized by a complex hydrogeological structure. During the circulation of underground waters dissolution of uranium from bedrocks in deep-lying rocks and its carryover into the soil formations located above could cause their enrichment with U-238. On the other hand, the opposite migration of Th-232 from the soil in bedrocks can cause, accordingly, its depletion. The action of these processes, depending on external factors, can cause observable deviations of the U-238/Th-232 ratio in both positive and negative directions from the average value.

Similar processes can also cause deviations of the Ra-226/U-238 ratio from equilibrium values. Ra isotope is easily leached and washed away by water: in natural formations Ra-226 often accumulates in quantities exceeding the equilibrium with uranium [4, 5]. The investigated region is

characterized by complex hydrogeological structure, and the effect of it and other geochemical factors can lead to appreciable variations in the activity ratios of these radionuclides, in particular, causing their observable deviations in the positive direction from equilibrium values.

The observed differences in the concentration of some families of radionuclides in samples of various soils can be related to features of the conditions of formation of genetic types of soil that can affect the content of natural radionuclide elements. A certain dependence of radionuclides on soil type was noted by Kogan *et al.* [4]; in particular, it was shown that the average content of naturally occurring radioactive elements decreases from primitive grey desert soils and grey-brown soils to sod-bog and bog soils.

The marked difference in the U-238/Th-232 ratio noted for certain soils (and to a certain degree, the Ra-226/U-238 ratio) is connected, apparently, with various characters of hydrological processes in foregoing Tbilisi water pressure hydrological system and in hydrological systems of Akhaltsikhe lava formations and the Javakheti ridge.

The high activity noted for sample 198 is apparently connected with effect of the rocks occurring at a shallow depth in this territory, outcrops of which were observed in this location. Therefore, the nearby selected schist sample (brecciated, light gray) has an equivalent activity of approximately 170 Bq/kg (data in preparation). Processes of physical hypergenesis of rocks, possible migration of their microparticles, and mixing with a soil layer could cause hyperactivity of the soil at a given local site.

Dependence on the type of geotectonic structure is not expressed very markedly, and can be connected with insufficient statistics. However, observable differences of activity probably can be connected with features of rocks in corresponding locations. Radioactivity data of rocks which are assumed to carry out in this region can represent certain interest under this problem.

A prominent feature recorded in the majority of samples regarding the activity ratio Pb-210/Ra-226 – marked excess of the equilibrium value – has been connected in previous studies with excess Pb-210 precipitation from the atmosphere. However, given the complex geological-tectonic structure of the investigated region, it is possible that the exhalation of radioactive radon gas from deep layers of rocks and soil could be the cause. Soil radon migrates through the upper soil layers (and further in atmospheric air), decays, and its decay products (including Pb-210) are deposited in the soil solid matter. This process can lead to the occurrence of a “nonequilibrium” concentration of Pb-210 (so-called “allochthonous” Pb-210 [31]).

The radionuclide Be-7, which is formed in the upper atmosphere as a result of interaction with space radiation and then combines with deposits in the soil, was detected in only one sample. Its absence from the other samples could be associated with the long period of samples storage, which could have led to a reduction in concentration to values below the minimal detectable level.

The distribution of the naturally occurring radionuclide K-

40 was similar to values observed by Kogan et al. [4].

Data for the technogenic radionuclide Cs-137 are of special interest. Cs-137 is generated in nuclear tests, which were regularly carried out in the past, and as a result, a background (equilibrium) concentration formed. According to various data, in particular, observations of the region of East Georgia [32] indicate that the Cs-137 background activity is now in the range of 1-10 Bq/kg. This level could be considered as the background value for the whole territory of Georgia connected with nuclear tests. However, there are also sites (for example, as a result of the Chernobyl accident) that have a much higher activity of technogenic radionuclides, in particular, Cs-137. For the last period, the activity of Cs-

137 has decreased considerably, due to washing away processes or migration into deeper layers. Variations in activity could also be due to non-uniform precipitations following the accident.

Some reference data from studies carried out in other regions of the world are cited in Table 7. The values in the current study were, on average, much lower than in other regions as well as compared to worldwide average values. It is also interesting that the value of radium equivalent activity, which varied from 48.6 to 93.1 Bq/kg, was below the recommended limiting value of 370 Bq/kg, while the annual effective dose (0.035-0.122 mSv/y) was below limiting value of 1 mSv/y [8, 9, 33].

Table 7. Activity concentration of radionuclides and other parameters in soil in various regions of the world.

SR	ST	Th-232	U-238	Ra-226	Pb-210	U-235	Be-7	K-40	Cs-137
Sp	UMIS	43-84	64-3388	81-11038	157-10190				
Pk		52.5		24.7				368	13.6
		43.4-62.4		20.0-29.4				163-493	1.3-54.1
	MS	57.0	52.2	36.8	25.8			1178	34.6
Nw		33.7-83.7	41.5-64.9	24.6-47.7	3.9-45.4			820-1320	2.3-85.1
	CS	39.3	39.0	18.7	29.7			890	63.4
		12.6-61.2	28.1-47.1	12.6-30.7	<M-118			652-1070	1.2-346
Ng	UnS	17.41		29.61				262.63	
		9.72-34.13		16.27-52.19				134.50-395.15	
Tk	UnS	26.9	18.0	22.4		1.40	2.11	419	253
Ar		3.8-44.9	8.5-37.3	8.3-41		0.6-2.6	0.9-6.8	197-950	0.5-13.4
Az		29-60	28-70	32-77				310-420	
		10-56	26-50	15-35				60-180	
Ge	GCD	24.7	29.7	34.8	55.0	1.4	-	455	13.6
		21.2-29.0	20.6-47.9	27.8-41.2	39.5-64.5	0.92-2.3		382-502	4.1-25.4
-"	Cn-Ld	50.5	24.9	28.9	66.5	1.2	42.6	479	12.7
		12.9-64.9	16.4-44.9	15.2-51.0	26.1-132	0.74-2.1	42.6-42.6	333-942	4.1-30.9
-"	Cn	28.2	26.9	31.9	39.8	1.3	30.2	575	5.7
		17.9-39.9	18.5-42.2	25.3-41.2	27.3-55.5	0.87-2.0	27.9-32.6	452-752	1.0-14.1
-"	Cz	26.5	18.4	25.1	65.4	0.86	-	358	16.7
		22.3-43.8	17.1-27.9	22.7-43.2	19.5-177	0.78-1.4		374-473	0.7-57.5
-"	Cn-Cr	21.2	21.3	29.3	79.0	1.0	-	511	36.5
		19.8-23.0	21.1-21.6	23.1-37.9	38.3-137	0.96-1.1		406-670	4.0-88.1
-"	BFWU	24.7	29.7	34.8	55.0	1.4	-	455	13.6
		21.2-29.0	20.6-47.9	27.8-41.2	39.5-64.5	0.92-2.3		382-502	4.1-25.4
Ww		45	33	32				412	

Table 7. Continued.

SR	ST	Ra _{eq}	Ra _{eq-al}	U-238/Th-232	Ra-226/U-238	Pb-210/ Ra-226	AEDE, mSv/y	AEDE _{al} mSv/y	Ref
Sp	UMIS								[6]
Pk		126.6					0.072		[7]
		107.3-147.8					0.061-0.084		
Nw	MS								[10]
	CS								
Ng	UnS	74.71							[11]
		51.04-100.85							
Tk	UnS								[12]
Ar									[34]
Az									-"
Ge	GCD	102	122	1.26	1.28	1.66	0.059	0.070	Pr.
		92.2-118	96.6-141	0.71-2.26	0.86-1.99	0.96-2.32	0.053-0.063	0.062-0.081	study
-"	Cn-Ld	102	139	0.99	1.17	2.40	0.059	0.080	-"
		57.1-210	67.8-262	0.63-1.54	0.91-1.47	1.25-5.75	0.036-0.122	0.042-0.152	-"
-"	Cn	112	120	1.01	1.28	1.32	0.066	0.070	-"
		83.6-148	87.8-137	0.53-1.34	0.92-2.05	0.70-2.20	0.048-0.092	0.053-0.086	-"

SR	ST	Ra _{eq}	Ra _{eq-al}	U-238/Th-232	Ra-226/U-238	Pb-210/ Ra-226	AEDE, mSv/y	AEDE _{al} mSv/y	Ref
"-	Cz	88.0	128	0.62	1.18	2.27	0.049	0.071	"-
		84.8-139	88.0-244	0.57-1.25	0.93-1.73	0.86-6.87	0.051-0.071	0.049-0.139	
"-	Cn-Cr	95.4	144	1.01	1.38	3.00	0.054	0.082	"-
		83.7-103	94.8-216	0.94-1.07	1.07-1.80	1.42-5.96	0.047-0.064	0.054-0.128	
"-	BFWU	78.9	126	0.59	1.44	3.43	0.044	0.071	"-
		61.4-96.4	97.0-155	0.53-0.64	1.34-1.54	3.22-3.65	0.035-0.053	0.055-0.086	
Ww									[34]

Note. 1) Studied regions: Sp – Spain; Pk – Pakistan; Nw – Norway; Ng – Nigeria; Tk – Turkey; Ar – Armenia; Az – Azerbaijan; Ge – Kvemo Kartli region, Georgia; Ww - Worldwide average values.

2) Soil types: UMIS – uranium mining-impacted soils; MS – moraine soil; CS – colluvium soil; UnS- undisturbed soil; GCD – grey cinnamonic dark; Cn-Ld – cinnamonic leached; Cn – cinnamonic; Cz – chernozem; Cn-Cr – cinnamonic calcareous; BFWU – brown forest weakly unsaturated.

5. Conclusion

Radioactivity of samples of soil of different types selected in different geotectonic zones and subzones of the Kvemo Kartli region was studied in the present work. Practically all radionuclides with marked gamma-radiation were identified. Generally, there are identified up to 22 radionuclides: the Th-232 family (Ac-228, Th-228, Ra-224, Pb-212, Bi-212, and Tl-208), the U-238 family (Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, and Pb-210), the U-235 family (U-235, Th-231, Th-227, Ra-223, Rn-219, and Pb-211), other naturally occurring radionuclides Be-7, K-40, and the techogenic radionuclide Cs-137. Radioactivity depending on the soil type and location was analyzed, and it was shown that radioactivity differs a little – the highest values of equivalent activity were observed for soil types GCD, Cn-Ld, and Cn, with average values from 102 to 112 Bq/kg and rather less for soils Cz, Cn-Cr, and BFWU, from 78.9 to 95.4 Bq/kg.

The geochemical activity ratios, in particular U-238/Th-232, Ra-226/U-238, were considered, and their marked deviations from the average and equilibrium values were established – it was noted that underground waters can be one of the possible reason. The ratio Pb-210/Ra-226 in almost all samples was appreciably greater than the equilibrium value (the greatest value of 6.87); certain correlation with features of geotectonic structure of studied territory was observed. Thus, it is possible to assume, that superequilibrium (allochthonous) Pb-210 can be connected not only with an atmospheric precipitation, but also with geotectonic faults.

The minimal and maximal values of annual effective dose differed by more than 3-fold and were in the range 0.035-0.122 mSv/y, respectively, and did not exceed the accepted permissible values.

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