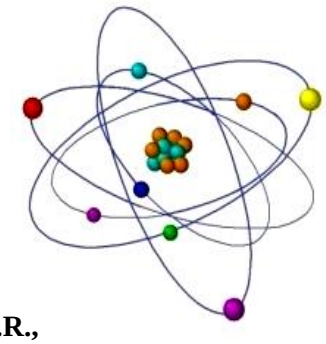
RADIOACTIVITY CONCENTRATION AND ASSESSMENTOF GAMMA-RADIATION EXPOSURE FROM THE SOIL OF DIFFERENT TYPE IN TERRITORY OF TBILISI CITY



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ABSTRACT

There are given results of radioactivity research in different types of soil within the territory of some districts of Tbilisi. Twenty samples of brown and alluvial types have been selected from 20 locations. Up to 22 radionuclides were identified. Concentration of radionuclides of Th-232 family was in limits from 16.8 to 41.4 Bq/kg (average value of 22.6 Bq/kg), U-238 family – from 17.7 to 67.8 Bq/kg (average value of 29.3 Bq/kg), U-235 family - from 1.09 to 3.76 Bq/kg (average value of 1.77 Bq/kg). Also individual radionuclides have been identified: K-40 – from 365 to 618 Bq/kg (average value of 465 Bq/kg), Cs-137 – from 1.5 to 118 Bq/kg (average value of 19.9 Bq/kg); trace amounts of Be-7 were determined in several samples. Radium equivalent activity varied from 71.5 to 148 Bq/kg (average value of 95.8 Bq/kg. Annual effective dose varied from 0.042 to 0.088 mSv/y (average value of 0.058 mSv/y). There were marked some features of radionuclides distribution, in particular, depending on the type of soil and sampling locations. Several 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.

Key words: radionuclides, soil, radioecological monitoring.

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. In many works the activity ratios of different 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, 4, 5].Therefore, it is important that such studies be carried out in each territorial region.

So, for example, in the work [3] there were studied soil samples selected in the territory of the Arnás River Catchment in the central part of the Spanish Pyrenees where six types of soil are widely distributed–Calcaric Fluvisols, Eutric Gleysols, Calcaric Regosols, Rendsic Leptosols, HaplicKastanozems and HaplicPhaeozems; it was determined activity concentration of Th-232, U-238, Ra-226, Pb-210, $Pb\text{-}210_{ex}$, K-40, Cs-137; correlation between soil radioactivity and its mineralogical content was investigated; values of activity ratios U-238/Ra-226 and Th-232/Ra-226 were calculated.

In another work [6] it was investigated activity of radionuclides Th-232, U-238, K-40, Cs-137 in samples, collected from uncultivated fields within the territory around Kestanbol (Çanakkale), Turkey;

there were calculated values of radium equivalent activity; authors noted that these values of the study area are higher than the limit value of 370 Bq/kg.

Similar researches of naturally occurring radioactive materials (NORM) - Th-232, U-238, Ra-226, U-235, K-40 - in anthrosol-type soil nearby Belgrade have been carried out in the work [7]. In another work [8] soil samples collected from undisturbed sites (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 equivalent were calculated; authors noted that all the linear fit of the measured parameters were significantly further away from unity (1) which shows that the concentration of NORM were mainly influenced by the oil exploration and production activities in the area and not from the geological constituent of the area.

The radiological quality of sand from the beaches along the coastlines in Ghana was studied in the work [9]. It was established that only naturally occurring radionuclides were identified in studied samples.

Activity concentration of Th-232, Ra-226 and K-40 in soil samples from the Fen Complex area (Norwegian county of Telemark) was determined in the work [10].

In Georgia regular researches of natural (and also technogenic) radioactivity were not actually carried out. Rather detailed researches of radioactivity in various environmental objects have been carried out in 1986, after failure on the Chernobyl atomic power station and, basically, concerned technogenic radionuclides [11, 12, 13]. In these works it has been shown, that during this period in the territory of the Western Georgia, basically in the strip adjoining to the sea the big concentrations of technogenic radionuclides were observed (in particular, Cs-137 concentration made from several hundreds to some thousands of Bq/kg). It is possible to note also works [14, 15] in which results of research of radiation condition of coast of water area of Black sea during later period are given, in particular, presence of 7 natural (Ac-228, Ra-226, Bi-214, Pb-214, Pb-212, Pb-210, K-40) and 1 technogenic radionuclide (Cs-137) has been fixed in soil in some areas of Adzharia (Batumi, Gonio, Sarpi, Chakvi, Kvariati). Some results of the last period are given in a study by Kekelidze et al [16]. Urushadze and Manakhov studied content of technogenic radionuclides Cs-137 and Sr-90 in different types of soil in the territory of Georgia [17].

In the present work there are given results of radioactivity research of different soil types within the territory of several districts of Tbilisi city – the largest city and the capital of Georgia.

MATERIALS AND METHODS

Study area

The territory of Tbilisi city and its suburb, which is located around contact of the Adjara-Trialetian folded zone and the Georgian block, represents the crossed mountain area within average watercourse of the River Mtkvari. Tbilisi occupies deep kettle-shaped valley which width changes from 3000 - 4000 m to 35 - 40 m. River Mtkvari crossing a city almost in meridional direction, divides it on two parts: more raised right-bank and considerably lowered left-bank. In the right-bank part of the city among the main forms of a relief it is necessary to allocate Teleti ridge, Tabori ridge, Sololaki ridge, mountain Mtatsminda, Tskneti height. Teleti and Tabori ridges are divided by Krtsanisi depression on which River Tabahmelastskali proceeds, between Tabori and Sololaki ridges locate deep gorge of River Tsavkisistskali. 20 soil samples (Table 1) were selected in this territory from 20 control points (nearby settlements and districts of city – Kiketi (Ky), Didgori (Dr), Kojori (Kj), Tabakhmela (Tx), Shindisi (Sx), Krtsanisi (Kx), Ortachala (Ot), ZemoPonichala (Zp), KvemoPonichala (Pl), University street (Un), Bagebi (Bb), Mtatsminda(Md), Sololaki (Sl), Botanic garden (Bg), Nakikala (Nr), Tabori (Ty). Types of selected samples are the following:

- cinnamonic – 16 samples (including cinnamonic calcareous (Cn-Cr) – 12 samples (220, 223, 229, 255, 264, 268, 272, 281, 286, 295, 303, 313), cinnamonic (Cn) – 4 samples (235, 238, 250, 253);
- alluvial – 4 samples (including alluvial calcareous (Al-Cr) – 4 samples (203, 204, 215, 217))

Table 1
List of control points (CP), field numbers (FN) of investigated samples and their types (ST)

#	CP	Lt(N); Ln(E)	FN	ST
1	Ky-2	41.64574; 44.64459	235	Cn
2	Dr-3	41.66658; 44.65155	238	-"-
3	Kj-7	41.65925; 44.69699	250	-"-
4	Tx-1	41.65475; 44.74512	253	-"-
5	Sx-2	41.67025; 44.76433	255	Cn-Cr
6	Kx-12	41.67191; 44.80206	229	-"-
7	Kx-3	41.66384; 44.80876	223	-"-
8	Kx-11	41.67016; 44.81483	220	-"-
9	Ot-2	41.67087; 44.83550	217	Al-Cr
10	Ot-3	41.66283; 44.87747	215	-"-
11	Zp-1	41.64281; 44.89925	204	-"-
12	Pl-1	41.63803; 44.93040	203	-"-
13	Tj-2	41.70367; 44.70393	295	Cn-Cr
14	Un-7	41.71841; 44.70726	313	-"-
15	Bb-8	41.70564; 44.73715	303	-"-
16	Md-2	41.69876; 44.79234	286	-"-
17	Sl-7	41.69055; 44.79087	264	-"-
18	Bg-6	41.68726; 44.80043	281	-"-
19	Nr-2	41.68791; 44.80956	272	-"-
20	Ty-2	41.68606; 44.81288	268	-"-

Note. Lt(N) – latitude (north); Ln(E) – longitude (east)

Sampling and analysis

Sampling

Samples were selected used the special hand auger directly in plastic containers (volume up to 2.0 L). After drying in laboratory conditions samples were grinded and sieved for their homogenization. Then samples were dried at the temperature 105 - 110°C to constant weight and their bulk density and weight were determined. These values were used at the description of sample geometry. The samples were sealed in Marinelli beaker (besides polyvinyl chloride adhesive tape was used also for hermetic sealing) and stored for more than 4 weeks to achieve secular equilibrium between Ra-226 and Rn-222.

Measurement of gamma radiation activity

Measurements were carried out using a Canberra GC2020 gamma spectrometer with a semi-conductor germanium detector with a relative efficiency 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 2.2% to 6.5%), U-238 (by the Th-232 line of 63.3 keV with an error in the diapason varying from 5.6% to 8.2%), Ra-226 and U-235 (by the 186 keV line, which was divided using the S506 program with error of 11.0%-24.4% for Ra-226, and 6.8%-12.0% for U-235), Pb-214 and Bi-214 (with error from 2.3% to 2.8%), Pb-210 (by the 46.54 keV line with an error from 9.4% to 20.4%). 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].

With an account taken of the influence of matrix composition, the chemical composition of samples were determined on the basis of reference data [18], 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 [19] 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 [20]:

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

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

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

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, 0.7×10^3mSv/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):

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

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

When allochthonousPb was present, the contribution of its "allochthonous" parents Pb-214 and Bi-214 (and, accordingly, the activity $Ra_{eq\text{-}al}$ 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 [21], 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 $0.985 \bullet A_{Ra\text{-}al}$was added into the calculation formula (1) where $A_{Ra\text{-}al}$ is equal to$A_{Pb\text{-}al} = A_{Pb} - A_{Ra}$ [3]. The similar term ($0.456 \bullet A_{Ra\text{-}al}$) 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 Bk/kg (equivalentto γ-radiations dose of 1.5 mSv/y) [22] 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 radioactivity[1] in the range from 300 to 1000 Bq/kg. The technique is described in more detail in works [23, 24].

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 material of the detector or the sample).The activity of identified radionuclides of the different families varied from 1.09 Bq/kg (for U-235 family) to 72.9 Bq/kg (for U-238 family). Among individual radionuclides K-40 had the greatest activity (up to 618 Bq/kg). The activity of several radionuclides 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 $Pb\text{-}210_{al}$ (Ra_{eq}) and taking this into account ($Ra_{eq\text{-}al}$) 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 2-5.

General characteristics

Within families of radionuclides, activity varied within sufficiently wide limits (Table 2). In particular, the range (mean) was 16.8-41.4 (22.6) Bq/kg for Th-232, 17.7-67.8 (29.3) Bq/kg for U-238, and 1.09-3.76 (1.77) Bq/kg for U-235. The activity of K-40 varied from 365 to 618 Bq/kg (mean, 4665). Trace amounts of radionuclide Be-7 was determined in several samples. Cs-137 was measured in all samples and ranged from 1.5 to 118 Bq/kg (mean, 19.9). The activity ratio U-238/U-235 (within ±10%) was 21.7 (accepted for natural objects). The ratios U-238/Th-232 and Ra-226/U-238 for majority of samples deviated (more than 10%) from the average value of 0.81 (for closed systems) – varied in the range 0.78-2.62 (1.35), and from the equilibrium value (1.00) – varied in the range 0.66-1.76 (1.04); the ratio Pb-210/Ra-226 for majority of samples was appreciably (more than 20%[2]) greater than the equilibrium value (the greatest value of 8.51). 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 (75.0%), by the level of equivalent activity, belonged to the group with low radioactivity, and a small proportion ofsamples (25.0%) belonged to the group with average values of equivalent activity (Table 5); it is worthy of note that if one considers allochthonous activity, the majority of samples (85.0%) were in the average group.

Table 2

Activity concentrations (Bq/kg) of families' radionuclides Th-232, U-238, Ra-226, Pb-210, and U-235, radionuclides K-40, and Cs-137, equivalent activities with no account taken of allochthonous $Pb\text{-}210_{al}$ (Ra_{eq}) and taking this into account ($Ra_{eq\text{-}al}$), 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.

#	CP	Th-232	U-238	Ra-226	Pb-214	Bi-214	Pb-210	U-235	K-40	Cs-137	$Pb\text{-}210_{al}$	Ra_{eq}	$Ra_{eq\text{-}al}$	U-238/U-235	U-238/Th-232	Ra-226/U-238	Pb-210/Ra-226	AEDE mSv/y	$AEDE_{al}$, mSv/y
1	Ky-2	19.4	28.0	27.6	26.6	25.9	78.3	1.85	494	6.5	50.7	89.8	140	21.9	1.87	0.76	2.84	0.060	0.089
2	Dr-3	20.0	25.0	24.2	25.8	24.6	37.8	1.23	418	2.6	13.6	82.0	95	22.0	1.36	0.89	1.56	0.052	0.059
3	Kj-7	18.1	23.4	26.2	24.6	24.4	48.6	1.77	445	3.3	22.4	83.2	105	22.3	1.31	1.10	1.86	0.050	0.062
4	Tx-1	22.9	34.9	34.2	37.5	31.1	75.3	1.89	450	19.5	41.0	98.4	139	22.0	1.33	1.13	2.2	0.057	0.080
5	Sx-2	41.4	47.7	49.4	48.5	46.8	77.6	2.73	562	18.5	28.3	148	176	21.5	1.20	0.99	1.57	0.088	0.104
6	Kx-12	25.6	67.8	72.9	71.6	70.5	74.9	3.76	451	15.4	2.0	141	143	21.9	2.62	1.09	1.03	0.080	0.081
7	Kx-3	21.3	20.2	17.1	20.9	20.3	58.2	1.29	415	19.7	41.1	76.7	117	22.3	1.22	0.66	3.4	0.052	0.075
8	Kx-11	22.8	37.5	44.8	38.3	37.5	56.2	2.16	404	4.9	11.5	106	117	22.4	1.66	1.18	1.26	0.059	0.066
9	Ot-2	19.2	20.1	26.2	21.8	21.2	114	1.71	422	21.0	88.0	83.2	170	21.2	0.78	1.76	4.35	0.044	0.094
10	Ot-3	19.2	17.7	19.5	18.2	17.3	40.8	1.30	430	18.2	21.2	77.2	98.1	21.1	0.91	1.11	2.09	0.046	0.058
11	Zp-1	21.8	33.5	35.9	34.6	34.2	51.6	1.97	414	1.5	15.6	96.0	111	21.7	1.69	0.98	1.44	0.058	0.067
12	Pl-1	20.3	23.7	27.6	24.0	24.1	47.7	1.30	422	8.7	20.1	86.1	106	21.2	1.03	1.32	1.73	0.049	0.060
13	Tj-2	16.8	18.3	20.4	19.1	18.4	20.7	1.09	385	2.8	0.24	71.5	71.7	21.5	1.07	1.14	1.01	0.042	0.043
14	Un-7	22.8	25.2	29.3	26.0	26.5	53.8	1.53	518	3.1	24.6	98.1	122	22.4	1.12	1.15	1.84	0.058	0.072
15	Bb-8	23.0	29.6	25.9	29.3	29.8	83.0	1.72	541	28.4	57.2	96.7	153	21.7	1.52	0.74	3.21	0.065	0.097
16	Md-2	24.0	25.5	28.8	25.6	24.3	60.2	1.52	556	1.5	31.4	102	133	21.3	1.12	1.07	2.09	0.062	0.079
17	Sl-7	20.7	31.9	31.2	33.1	31.9	37.0	1.82	520	5.1	5.8	97.2	103	22.3	1.88	0.80	1.18	0.064	0.067
18	Bg-6	26.1	29.0	33.2	30.1	28.8	223	2.05	618	118	190	114	301	22.3	1.09	1.17	6.73	0.067	0.174
19	Nr-2	19.6	22.3	18.6	24.1	23.6	103	1.32	365	62.1	84	72.1	155	20.8	1.32	0.72	5.51	0.048	0.095
20	Ty-2	27.3	24.3	26.4	24.5	24.5	224	1.41	468	36.9	198	98.2	293	21.3	0.91	1.06	8.51	0.058	0.169
	av	*22.6*	*29.3*	*31.0*	*30.2*	*29.3*	*78.3*	*1.77*	*465*	*19.9*	*47.3*	*95.8*	*142*	*21.8*	*1.35*	*1.04*	*2.77*	*0.058*	*0.085*
	mn	*16.8*	*17.7*	*17.1*	*18.2*	*17.3*	*20.7*	*1.09*	*365*	*1.5*	*0.24*	*71.5*	*71.7*	*20.8*	*0.78*	*0.66*	*1.01*	*0.042*	*0.043*
	mx	*41.4*	*67.8*	*72.9*	*71.6*	*70.5*	*224*	*3.76*	*618*	*118*	*198*	*148*	*301*	*22.4*	*2.62*	*1.76*	*8.51*	*0.088*	*0.174*

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	Th-232			U-238			Ra-226			Pb-210			U-235			K-40			Cs-137		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	Cn	20.1	18.1	22.9	27.8	23.4	34.9	28.0	24.2	34.2	60.0	37.8	78.3	1.69	1.23	1.89	452	418	494	8.0	2.6	19.5
2	Cn-Cr	24.3	16.8	41.4	31.6	18.3	67.8	33.2	17.1	72.9	89.3	20.7	224	1.84	1.09	3.76	484	365	618	26.3	1.5	118
3	Al-Cr	20.1	19.2	21.8	23.8	17.7	33.5	27.3	19.5	35.9	63.5	40.8	114	1.65	1.32	2.05	422	414	430	12.4	1.5	21.0

Table 4

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

№	ST	Ra_{eq}			$Ra_{eq\text{-}al}$			U-238/Th-232			Ra-226/U-238			Pb-210/Ra-226			AEDE, mSv/y			$AEDE_{al}$, mSv/y		
		av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx	av	mn	mx
1	Cn	88.4	82.0	98.4	120	95.4	140	1.47	1.31	1.87	0.97	0.76	1.13	2.11	1.56	2.84	0.055	0.050	0.060	0.073	0.059	0.089
2	Cn-Cr	102	71.5	148	157	71.7	301	1.39	0.91	2.62	0.98	0.66	1.18	3.11	1.01	8.51	0.062	0.042	0.088	0.093	0.043	0.174
3	Al-Cr	85.6	77.2	96.0	121	98.1	170	1.10	0.78	1.69	1.29	0.98	1.76	2.40	1.44	4.35	0.049	0.044	0.058	0.070	0.058	0.094

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

Table5

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

#	GA	$A_{eq,}$ (Bq/kg)	A_{av}, (Bq/kg)	N_s	r (%)
1	II	30 - 100	87.1	15	75.0
2	III	100 - 300	122	5	25.0

DependenceontheType

The highest values of equivalent activity (Table 3, Table 4) were observed for soil type Cn-Cr, with average value of 102 Bq/kg,

and rather less for soils Cn and Al-Cr – 85.6 and 88.4 Bq/kg. The activity ratio U-238/Th-232 for soils of all types GCD exceeded (within ±10%) the average value of 0.81. The ratio Ra-226/U-238 in all soil types met (within ±10%) or exceeded the equilibrium value. The highest excess values for ratio Pb-210/Ra-226 were determined for Cn-Cr soil.

Radiological Parameters

Determined minimal and maximal values of annual effective dose varied in the range 0.042-0.088 mSv/y (Table 2). These values (as well as equivalent activity) increase (annual effective dose - in the range 0.043-0.174 mSv/y) at the assumption, that allochthonous Pb-210 is caused by excess soil radon.

DISCUSSION

Concentration of radioactive elements in soils is determined by radioactivity of initial rocks and set of the subsequent soil formation processes. Content and concentration of naturally occurring radionuclides, in general, correspond to those usually observed for different soils [4]. In soil samples due to specific processes of their formation (the big role of migratory processes therefore hashing of various minerals occurs much more effectively, than in rocks) range of activity changes of natural radionuclides in them is much less, than in rocks (where they are in the "sealed" condition). Owing to these reasons also, as it is apparent from results, it is not observed the noticeable expressed dependence on soil type. Certain interest represents disbalance between Ra-226 and Pb-210 observed in the work, in particular, high values of the ratio Pb-210/Ra/226. In a number of works presence of "superequilibrium" (allochthonous) Pb-210 is noticed that connects with radon migration from the bottom layers of earth in the top (and further in atmosphere). So, in the work [25]it is noticed, that Pb-210 is capable to accumulate on the walls of pores and faults at the passage of radon flow through soil. At the passage of radon through a layer from underlying horizons there is an accumulation in a layer of excess Pb-210, not supported by radium, formed by the expense of decay of radon arriving from below. Radioactive balance in soil is thus broken towards increase in activity of Pb-210, i.e. ratio Pb-210/Ra-226 > 1 is realized. Activity ratio Pb-210/Ra-226 in soils is the integrated characteristic of existence in system of a constant or pulse ascending radon flow for a long time. In a number of works presence of "superequilibrium" Pb-210 in soil is connected with its deposition from atmospheric air. Prominent feature of activity ratio Pb-210/Ra-226 observed in the samples, in particular, considerable excess of equilibrium value of activity size in the majority of samples - apparently denotes appreciable effect of the factor of radioactive gas radon exhalation from deep soil layers or subsoil rocks. It is possible to assume, that considerably raised values of ratio Pb-210/Ra-226 can serve as the certain identifier of the territory with the raised seismic dangerous.

This question represents certain interest and the further researches in this direction are necessary. The question of the nature of nonequilibrium Pb-210 is of interest also from the radiological point of view. 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 trace amounts in several samples. 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.

Data for the technogenic radionuclide Cs-137 are of special interest. As it is apparent from results, it was observed in all samples in sufficiently appreciable amounts. Usually its occurrence is connected with failure of the Chernobyl atomic power station in 1986. By a number of data, in particular, according to systematic observations for the flat areas of East Georgia [26], values of Cs-137 activity are now, basically, in the range 1 - 10 Bq/kg. With certain degree of convention it is possible to consider this level as background value for the whole territory of Georgia. Average value (19.9 Bq/kg) is greater this quantity that can be due to non-uniform precipitations following the accident. However it is impossible to exclude completely that the pollution fact could have rather recent history, considering presence of nuclear objects in surrounding geographical region. Results received for location Bg-6 (see Table 2) are of the special interest that identifies necessity of additional researches for these places.

The distribution of the naturally occurring radionuclide K-40 was similar to values observed by Kogan et al. [4].

The calculated values of annual effective dose do not exceed 1 mSv/y dose limit recommended for public radiation exposure control [27, 28, 29].

Some reference data from studies carried out in other regions of the world are cited in Table 6. The values in the current study were, on average, much lower than in other regions as well as compared to worldwide average values. In conclusion it is necessary to note, that the received results represent doubtless scientific and applied interest for the investigated region that confirms an urgency of such researches and necessity of their regular character.

Table6

Activity concentration of radionuclides in soil in different regions of the world

SR	ST	Th-232	U-238	Ra-226	Pb-210	U-235	K-40	Cs-137	Ra_{eq}	$Ra_{eq\text{-}al}$	U-238/ Th-232	Ra-226/ U-238	Pb-210/ Ra-226	AEDE, mSv/y	$AEDE_{al}$ mSv/y	Ref
Tk	UnS	192 151.91-275.63	115 82.32-166.99				1207 1015.48-1484.93	18.1 0.37-36.03	498 430-626							[6]
Nr		3280–8395		89–171			404–654									[10]
Gh	Bs	42.6 16.8-231.2	20.08 11.1-31.8	31.4 10.9-103.7		0.91 0.51-1.5	110 68.3-184		101.0 59.5-445		0.79 0.11-1.56	1.27 0.42-4.17		0.066 0.037-0.251		[9]
Sp		34.6 23.7-49.4	40.2 19.9-60.0	26.7 20.8-34.9	74.0 26.7-140.0		586.2 446-799	30.9 4.4-64.7								[3]
Ng	UnS	17.41 9.72-34.13		29.61 16.27-52.19			263 135-395		74.7 51.0-101							[8]
Sb	An	53.1 45-62	67.4 49-90	48.1 39-59		3.6 2.7-4.6	642 565-755									[7]
Ar		29-60[3]	28-70[3]	32-77			310-420									[30]
Az		10-56[3]	26-50[3]	15-35			60-180									-"-
Ww		45[3]	33[3]	32			412									-"-
Ge	Cn	20.1 18.1-22.9	27.8 23.4-34.9	28.0 24.2-34.2	60.0 37.8-78.3	1.69 1.23-1.89	452 418-494	8.0 2.6-19.5	88.4 82.0-98.4	120 95.4-140	1.47 1.31-1.87	0.97 0.76-1.13	2.11 1.56-2.84	0.055 0.050-0.060	0.073 0.059-0.089	Pr. study
-"-	Cn-Cr	24.3 16.8-41.4	31.6 18.3-67.8	33.2 17.1-72.9	89.3 20.7-224	1.84 1.09-3.76	484 365-618	26.3 1.5-118	102 71.5-148	157 71.7-301	1.39 0.91-2.62	0.98 0.66-1.18	3.11 1.01-8.51	0.062 0.042-0.088	0.093 0.043-0.174	-"-
-"-	Al-Cr	20.1 19.2-21.8	23.8 17.7-33.5	27.3 19.5-35.9	63.5 40.8-114	1.65 1.32-2.05	422 414-439	12.4 1.5-21.0	85.6 77.2-96.0	121 98.1-170	1.10 0.78-1.69	1.29 0.98-1.76	2.40 1.44-4.35	0.049 0.044-0.058	0.070 0.058-0.094	-"-

Note. 1) Studied regions (SR): Tk – Turkey; Nr – Norway; Gh – Ghana; Sp – Spain; Ng – Nigeria; Sb – Serbia; Ar – Armenia; Az – Azerbaijan; Ww - Worldwide average values; Ge – Tbilisi, Georgia.

2) Soil types: UnS- uncultivated, undisturbed soil; Bs – beach sand; An – anthrosol-type soil; Al-Cr – alluvial calcareous; Cn – cinnamonic; Cn-Cr – cinnamonic calcareous.
3) Values are given for Th-232 and U-238, correspondingly.

CONCLUSION

1. Up to 22 radionuclides are identified in soil samples selected from the 20 locations in the territory of several districts of Tbilisi city: Th-232 family – Ac-228, Th-228, Ra-224, Pb-212, Bi-212, Tl-208; U-238 family – Th-234, Pa-234, Th-230, Ra-226, Pb-214, Bi-214, Pb-210; U-235 family – U-235, Th-231, Th-227, Ra-223, Rn-219, Pb-211; other naturally occurring radionuclides – Be-7, K-40, and also technogenic radionuclide Cs-137
2. Activity of naturally occurring radionuclides depending on the soil type and location differs a little – the highest values of equivalent activity were observed for samples of cinnamonic calcareous soil (average value of 102 Bq/kg), and rather less for samples of cinnamonic and alluvial calcareous soils (average values 85.6 and 88.4 Bq/kg, respectively).
3. Several activity ratios, in particular, U-238/U-235, U-238/Th-232, Ra-226/U-238 and Pb-210/Ra-226 have been considered, and some features in their distribution have been established; it was shown that for ratio Pb-210/Ra-226 it is observed considerable deviations in the greater way from equilibrium value.
3. Several radiological parameters, in particular, annual effective doses were calculated; it was shown that these values are lowering in comparison with recommended norms.
4. Comparison with reference data as well as analysis of obtained results and some of their features was carried out.

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REFERENCES

1. G. Faure, Principles of Isotope Geology, second ed., Wiley, New York, 1986.

2. UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), Exposure from Natural Sources of Radiation, 1993 Report to the General Assembly, Annex A, United Nations, New York, 1993.

3. A. Navas, L. Gaspar, M.López-Vicente, J. Machín, Spatial distribution of natural and artificial radionuclides at the catchment scale (South Central Pyrenees), Radiat. Meas. 46 (2011) 261-269, http://dx.doi.org/10.1016/j.radmeas.2010.11.008.

4. R.M. Kogan, I.M. Nazarov, Sh.D. Fridman, Basics of Environmental Gamma-Spectrometry, Atomizdat, Moscow, 1976 [in Russian].
5. N.A. Titaeva, Nuclear Geochemistry, MGU, Moscow, 2000 [in Russian].
6. B. Merdanoğlu, N. Altinsoy, Radioactivity concentrations and dose assessment for soil samples from Kestanbol granite area, Turkey, Radiat. Prot. Dosim. 121 (4) (2006) 399-405, http://dx.doi.org/10.1093/rpd/ncl055.
7. I. Vukašinović, A. Đorđević, M.B. Rajković, D. Todorović, V.B. Pavlović, Distribution of natural radionuclides in anthrosol-type soil, Turk. J. Agric. For., 34 (2010) 539-546. DOI:10.3906/tar-0911-59.

8. G.O. Avwiri, C.P. Ononugbo, Natural radioactivity levels in surface soil of Ogba/Egbema/Ndoni oil and gas fields, Energy Science and Technology, 4 (2) (2012) 92-101. DOI:10.3968/j.est.1923847920120402.427.
9. H. Lawluvi, E.O. Darko, C. Schandorf, A. Fannu, A.R. Awudu, D.O. Kpeglo, Natural radioactivity concentrations in beach sands from some tourists resorts, Res. J. of Env. and Earth Sciences, 3(6) (2011) 729-736.
10. J. Mrdakovic Popic, Ch.R. Bhatt, B.t Salbu, L. Skipperud, Outdoor 220Rn, 222Rn and terrestrial gamma radiation levels: investigation study in the thorium rich Fen Complex, Norway, J. Environ. Monit., 14 (2012) 193-201.
11. K.Sh. Nadareishvili, M.S. Tsitskishvili, G.A. Gachechiladze, N.M. Katamadze, L.N. Intskirveli, S.R. Kirtadze, D.N. Mandzhgaladze, L.M. Mosulishvili, T.G. Sanaya, R.E. Hazaradze, R.D. Chitanava, N.N. Shavdiya, Effect of Chernobyl accident on radio ecological situation in the Caucasus. Paper 1: Radionuclide echo of Chernobyl in Georgia,Radiat. Stud., 6 (1991) 132-151 [in Russian].
12. L.M. Mosulishvili, N.I. Shonia, N.M. Katamadze, E.I. Ginturi, Radionuclides of Chernobyl etiology in the Republic of Georgia – kinetics of their accumulation and migration, Radiat. Stud., 6 (1994) 252-262 [in Georgian].
[13] N. Katamadze, L. Mosulishvili, N. Kuchava, D. Eristavi, N. Shonia, Dose of external irradiation of the population in Tbilisi region after Chernobyl accident, Radiat. Stud., 7 (1994) 263-272 [in Georgian].
14. S.V. Pagava, The study of radiation condition in coastal zone of Black Sea in the Chakvi-Sarpi region and adjacent water area, Soros educational journal, Ecology, 1 (2003) 53-62.
15. T. Museliani, J. Oniani, T. Oniani, Pollution of Black Sea coastal waters on the territory of the West Georgia, Bulletin of the Georgian Academy of Sciences, 171 (1) (2005) 180-181
16. N. Kekelidze, T. Jakhutashvili, B. Tutberidze, E. Tulashvili, M. Akhalkatsishvili, L. Mtsariashvili, Radioactivity of soils in Mtskheta-Mtianeti region (Georgia). Annals of Agrarian Science, 15 (2017) 304-311, http://dx.doi.org/10.1016/j.aasci.2017.07.003.
17. T.F. Urushadze, D.V. Manakhov, Radioactive contamination of the soils of Georgia, Annals of Agrarian Science,15 (2017) 375-379, http://dx.doi.org/10.1016/j.aasci.2017.07.009.
18. T. Urushadze, The main soils of Georgia, Metsniereba, Tbilisi, 1997.
19. National Nuclear Data Center, Brookhaven National Laboratory, USA, http://www.nndc.bnl.gov/nudat2/.
20. UNSCEAR (United Nations Scientific Committee on the Effect of Atomic Radiation), Exposure from natural radiation sources, 2000 Report to the General Assembly, United Nations, New York, 2000.
21. K. Saito, P. Jacob, Gamma ray fields in the air due to sources in the ground, Radiat. Prot. Dosim. 58(1) (1995) 29-45.
22. OECD (Organization for Economic Cooperation and Development), Exposure to Radiation from the Natural Radioactivity in Building Materials, Report by a Group of Experts of the OECD, Nuclear Energy Agency, Paris, France, 1979.
23. N. Kekelidze, T. Jakhutashvili, B. Tutberidze, E. Tulashvili, M. Akhalkatsishvili, L. Mtsariashvili, Radionuclides in rocks of southern part of Mtskheta-Mtianeti region (Georgia), Journal of Geochemical Exploration, 190 (2018) 1-9, https://doi.org/10.1016/j.gexplo.2018.02.010.
24. N.P. Kekelidze, T.V. Jakhutashvili, B.D. Tutberidze, E.V. Tulashvili, M.R. Akhalkatsishvi, L.A. Mtsariashvili, Radioactivity of rock samples of different origin (the central region of the Main Caucasian Range, Georgia), Science and Engineering Applications, 2 (2017) 181-192, https://doi:10.26705/SAEA.2017.2.14.181-192.

25. P.S. Miklyaev, Scientific Bases of a Radon-danger Estimation of Platform Territories, PhD Thesis, Institute of Geoecology, Moscow, 2015.
26. NATO-OSCE Project SfP 977991 “South Caucasus River Monitoring”, NATO Science for Peace Programme (2002-2008). www.kura-aks-natosfp.org.
27. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). 1988. Sources, effects and risks of ionizing radiation. Report to the General Assembly. United Nations, New York.
28. International Commission on Radiological Protection (ICRP). 2007. Publication 103, Ann. ICRP 37. Oxford, Pergamon.
29. Lj. Janković Mandić, and S. Dragović. 2010. Assessment of terrestrial gamma exposure to the population of Belgrade (Serbia). Radiat. Prot. Dosim. 140 (4):369–377.
30. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation. UNSCEAR 2008 Report, Annex B, Exposures of the public and workers from various sources of radiation, New York, United Nations, 2010.