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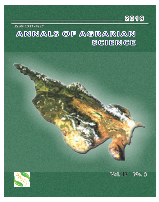

Radioactivity in soil samples from the settlements of Tbilisi city (the capital of Georgia)

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A B S T R A C T

This study investigated the radioactivity of various types of soil (cinnamonic, cinnamonic calcareous, grey cinnamonic and alluvial calcareous) in suburbs of Tbilisi, Georgia. Soil samples were collected from 11 locations and analyzed using gamma spectrometry. ^{232}Th varied in the range 16.9-34.2 Bq/kg (average 23.8 Bq/kg), ^{238}U – in the range 19.3-50.5 Bq/kg (27.4), and ^{235}U - in the range 0.88-2.3 Bq/kg (1.3). ^{40}K ranged from 291 to 560 Bq/kg (442). Technogenic radionuclide ^{137}Cs was identified in almost all samples and its activity concentration ranged from 3.6 to 27.6 Bq/kg (12.7). Some activity ratios were calculated: ^{238}U/^{235}U, ^{238}U/^{232}Th, ^{226}Ra/^{238}U and ^{210}Pb/^{226}Ra. The radium equivalent activity ranged from 66.1 to 136 Bq/kg (88.7). Annual effective gamma-dose rate varied in the range 0.046-0.083 mSv/y (0.056). Some features of radionuclide distribution are noted, and a comparison was carried out with existing data from the literary.

Keywords: Radioactivity, Gamma spectrometry, Radium equivalent, Gamma-dose, Natural radioactivity, Technogenic radioactivity.

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Introduction

The natural and technogenic radioactivity of soil is one of the main components of the radioactive background of the Earth. Numerous studies have shown that this radioactivity differs considerably in various areas of the globe, ranging from single units up to hundreds of Bq/kg.

Many studies also take into account the activity ratios of various radionuclides, in particular, ^{238}U/^{235}U, ^{238}U/^{232}Th, ^{226}Ra/^{238}U and ^{210}Pb/^{226}Ra as these help understand the various geochemical processes taking place in the rocks and soils [1-4]. Such researches are important for each region.

Soil of various types were studied in different countries [5-9]. Activity concentration of different naturally occurring radionuclides were investigated in these studies. Various activity ratios of the radionuclides were computed by the obtained results.

Here there are presented results of research on the radioactivity of samples of soil collected from 11 locations in the peripheral part of the largest city of Georgia, Tbilisi, and also from 4 locations in the suburbs of the nearby city of Mtskheta.

Materials and Methods

Study area. The territory of Tbilisi is characterized by a complex soil-geological structure. It occupies the eastern end of the Adzharia-Trialeti mountain system and is situated in the territory (Tr) of so-called Tbilisi artesian basin (**I**) – a water pressure system of fissure and fissure-karst waters. Some characteristics of surface waters in this territory, in particular, radon content, have been investigated previously [10], but the radioactivity of soil structures in this area has not been studied. For comparison soil samples from a nearby territory (approximately 15-20 km to the north of Tbilisi, in the territory of the Kartli artesian basin (**II**) were also studied. Interstitial, fissure and fissure-karst waters can also be found in this basin.

26 samples were collected in territory **I** from 11 locations (2-3 samples per location) near various settlements and sites – Tskneti (Ts), Kojori (Kj), Lilo (Ll), Zahesi (Zh), Avchala (Av), Betania (Bt), Lisi (Ls), and Sololaki (Sl), and 9 samples were collected in territory **II** from 4 locations – settlements Natakhtari (Nt), Misaktsieli (Ms), Bulachauri (Bl), and Bodorna (Bd), in particular, in territory **I**: alluvial calcareous (Al-Cr) – 6 samples; cinnamonic calcareous (Cn-Cr) – 9 samples; cinnamonic (Cn) – 8 samples; grey cinnamonic (GC) – 3 samples; in territory **II**: alluvial calcareous (Al-Cr) – 9 samples (Fig., Table 1).

Sampling. Samples were collected from a deep 0.2-0.4 m. After drying in laboratory conditions samples were ground and dried at 105-110^{0}C, and then sealed in Marinelli beaker and stored for more than 4 weeks.

Measurement of gamma radiation activity. Measurements were carried out using gamma spectrometer Canberra GC2020 with semi-conductor germanium detector with relative efficiency 24%. Gamma spectra acquisition time was 72 h. For the analysis it was used software Genie-2000. There were determined ^{238}U (^{234}Th), ^{226}Ra, ^{214}Pb and ^{214}Bi, ^{210}Pb, ^{235}U, the main radionuclides of ^{232}Th family and others (^{7}Be, ^{40}K, ^{137}Cs). Some geochemical ratios were also determined, in particular, the activity ratios ^{238}U/^{232}Th, ^{226}Ra/^{238}U and ^{210}Pb/^{226}Ra. More detailed description is given in the work [11]. After activity determination for each sample their averaging for each location was carried out.

Assessment of radium equivalent activity Ra_{eq} (Bq/kg), absorbed dose rate *D* (nGy/h) and annual effective dose equivalent AEDE (mSv/y) was carried out according to widely-used formulas [12].

Results

Up to 22 radionuclides were identified: in detail, ^{232}Th family – ^{228}Ac, ^{228}Th, ^{224}Ra, ^{212}Pb, ^{212}Bi, ^{208}Tl (six radionuclides), ^{238}U family – ^{234}Th, ^{234}Pa, ^{230}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{210}Pb (seven radionuclides), ^{235}U family – ^{235}U, ^{231}Th, ^{227}Th, ^{223}Ra, ^{219}Rn, ^{211}Pb (six radionuclides), and the individual radionuclides ^{7}Be and ^{40}K and the technogenic radionuclide ^{137}Cs.

Tables 2–3 present a number of radionuclide parameters: activity concentration (A), radium equivalent activity (Ra_{eq}), activity ratios of radionuclides ^{238}U/^{235}U, ^{238}U/^{232}Th, ^{226}Ra/^{238}U, ^{210}Pb/^{226}Ra, absorbed dose rate (*D*), annual effective dose equivalent (AEDE), and generalized data – average (av), minimal (mn) and maximal (mx) values – of activity concentration for study samples.

General characteristics. The activity of radionuclides of families varied over a relatively short range: ^{232}Th – from 16.9 to 34.2 Bq/kg (average 23.8 Bq/kg); ^{238}U – from 19.3 to 50.5 Bq/kg (27.4);

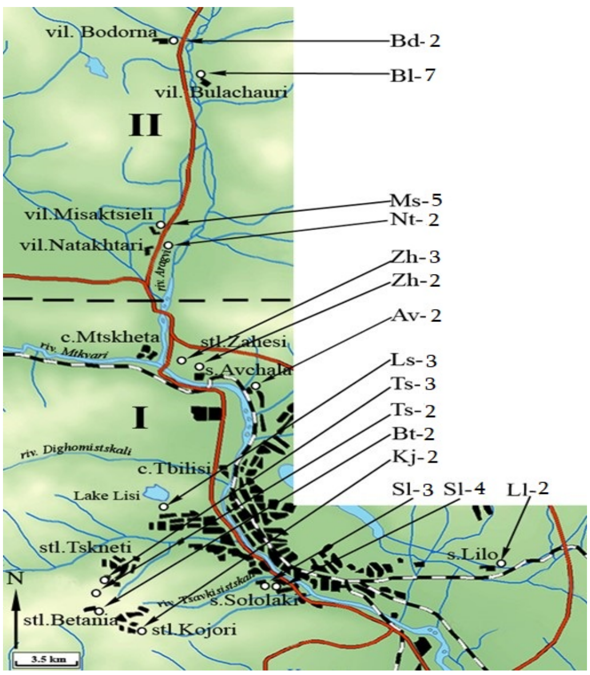

Fig 1. *Objects for investigation*

Table 1. *List of locations (L) and sample types (ST)*

#	Tr	L	Lt(N)[1]; Ln(E)[2]	ST
1	**I**	Ll-2	41.69241; 44.98823	GC
2	-“-	Sl-4	41.68834; 44.79035	Cn-Cr
3	-“-	Sl-3	41.68830; 44.79018	Cn-Cr
4	-“-	Kj-2	41.66133; 44.70335	Cn
5	-“-	Bt-2	41.66809; 44.65187	Cn
6	-“-	Ts-2	41.68028; 44.69153	Cn-Cr
7	-“-	Ts-3	41.68052; 44.69327	Cn-Cr
8	-“-	Ls-3	41.73970; 44.74306	Cn-Cr
9	-“-	Av-2	41.81945; 44.78584	Al-Cr
10	-“-	Zh-2	41.82641; 44.75507	Al-Cr
11	-“-	Zh-3	41.82860; 44.74047	Al-Cr
12	**II**	Nt-2	42.92344; 44.73327	Al-Cr
13	-“-	Ms-5	41.94054; 44.73627	Al-Cr
14	-“-	Bl-7	42.00925; 44.75929	Al-Cr
15	-“-	Bd-2	42.05653; 44.74404	Al-Cr

[1]Lt(N) – latitude (north); [2]Ln(E) – longitude (east).

^{235}U – from 0.88 to 2.3 Bq/kg (1.3) – more than 10 times less in comparison with ^{232}Th and ^{238}U; equivalent activity varied from 66.1 to 136 Bq/kg (88.7).

Note: sample Sl-3 showed relatively high activity (it was collected from a mountain slope) which considerably exceeded (more than half as much again) the activity of sample Sl-4, which was collected from a nearby point, as well as other samples.

Be was observed only in one sample (Kj-2); in other samples values were less than Minimal Detectable Activity (MA) or it was not measured. Activity of ^{40}K was the highest among all radionuclides – from 291 to 560 Bq/kg (442). Technogenic radionuclide ^{137}Cs was measured in almost all samples – from 3.6 to 27.6 Bqkg (12.7). All calculated values of the $^{238}U/^{235}U$ ratio corresponded (within 10%) to 21.7 (accepted for natural objects). Activities ratio $^{238}U/^{232}Th$ deviated (more than ±10%) from the average value of 0.81 (for closed systems) basically towards higher values (maximal value of 1.77 for sample Sl-4). For $^{226}Ra/^{238}U$ ratio appreciable (more than ±10%) prevalence of the daughter product was observed in five samples (Zh-2, Ts-2, Kj-2, Sl-3 and Ll-2), and that of the parent radionuclide in three samples (Ls-3, Bt-2 and Bl-7). For $^{210}Pb/^{226}Ra$ ratio six samples (Ls-3, Bt-2, Kj-2, Nt-2, Bl-7, Bd-2) showed appreciable prevalence (more than ±20% - the range of limits is expanded because the determination error of Pb-210 reached up to 20%) of the daughter product (the highest value was 2.47); one sample (Sl-3) showed prevalence of the parent radionuclide. The highest values of Ra_{eq}, D and AEDE were recorded in the sample collected from location Sl-3, and the least in the sample collected from location Ls-3.

Dependence on the type. The results showed no appreciable dependence on soil type (Table 3). The highest values of Ra_{eq} in territory **I** were found in samples of type Al-Cr – average value of 93.5 Bq/kg (which is sufficiently close to the average value of 95.1 Bq/kg for the samples of the same type in territory **II**); lower values were found for samples of type Cn-Cr (88.0 Bq/kg) and the lowest values were observed for soils of types Cn and GC (75.2 and 79.5 Bq/kg). There was a different picture for the $^{238}U/^{232}Th$ ratio: almost all samples from territory **I** had a ratio that considerably exceeded the average ratio of 0.81 while soil from territory **II** had a similar ratio to the average. The $^{226}Ra/^{238}U$ ratio exceeded a little equilibrium value in Al-Cr and GC soils and it was similar to this value in Cn-Cr and Cn soils in territory **I** and in Al-Cr soil in territory **II.** The $^{210}Pb/^{226}Ra$ ratio was close to the equilibrium value in Al-Cr, Cn-Cr and GC soils and exceeded the equilibrium value in Cn-Cr soil in territory **I** and in Al-Cr soil in territory **II.**

Radiological parameters. The minimal and maximal values of annual effective dose differed between points almost two fold, ranging from 46 to 83 µSv/y (Table 3). The lowest average values (46 µSv/y) were observed for soils of type GC, and the highest (59 µSv/y) for soils of type Al-Cr.

Table 2. *Some main radionuclides parameters of studied soil samples (designations are in the text)*

#	Tr	ST	L	^{232}Th	^{238}U	^{226}Ra	^{210}Pb	^{235}U	^{7}Be	^{40}K	^{137}Cs	Ra_{eq}	$^{238}U/^{235}U$	$^{238}U/^{232}Th$	$^{226}Ra/^{238}U$	$^{210}Pb/^{226}Ra$	D, nGy/h	AEDE, µSv/y
1	I	Al-Cr	Av-2	19.8	27.6	27.3	33.2	1.3	—	540	<M	89.5	21.3	1.40	0.99	1.22	47.2	58.1
2		-“-	Zh-2	28.8	21.3	31.1	26.9	0.99	-	521	12.2	98.2	21.5	0.74	1.57	0.87	48.9	60.2
3		-“-	Zh-3	22.8	35.3	36.4	29.7	1.6	-	456	20.5	92.9	21.6	1.55	1.03	0.82	49.1	60.4
4		Cn-Cr	Ls-3	16.9	26.4	14.0	27.2	1.2	—	354	9.3	66.1	21.3	1.56	0.53	1.94	37.1	45.7
5		Cn	Bt-2	21.7	24.4	19.8	28.4	1.1	-	425	5.8	78.4	21.7	1.12	0.84	1.42	42.1	51.7
6		Cn-Cr	Ts-2	19.6	19.3	27.3	29.3	0.88	—	403	22.8	73.7	21.8	0.98	1.42	1.07	37.5	46.2
7		-“-	Ts-3	20.4	28.9	25.1	25.7	1.3	-	469	10.0	83.6	21.4	1.41	0.88	1.01	45.3	55.7
8		Cn	Kj-2	17.6	21.9	27.3	46.3	1.02	6.1	418	27.6	71.9	21.6	1.24	1.32	1.71	38.1	46.8
9		Cn-Cr	Sl-3	34.2	50.5	60.9	41.1	2.3	—	559	3.6	136	21.9	1.48	1.21	0.67	67.3	82.8
10		-“-	Sl-4	18.7	33.1	30.9	<M	1.6	<MA	291	<MA	80.3	21.0	1.77	0.93	-	38.8	47.7
11		GC	Ll-2	19.3	25.6	42.2	37.0	1.2	-	345	14.9	79.5	21.5	1.32	1.66	0.98	37.8	46.5
1	II	Al-Cr	Nt-2	33.9	25.5	27.8	24.0	1.2	-	560	14.4	111	21.5	0.75	0.90	1.32	55.6	68.4
2		-“-	Ms-5	31.6	27.0	23.8	26.2	1.2	-	460	5.8	100	22.0	0.87	0.90	1.12	50.7	62.4
3		-“-	Bl-7	23.4	22.2	19.0	46.9	1.03	<MA	370	8.1	77.2	21.5	0.95	0.86	2.47	39.8	48.9
4		-“-	Bd-2	28.5	22.9	25.1	56.6	1.1	-	453	9.6	92.5	21.7	0.81	1.10	2.37	46.7	57.4
			av	*23.8*	*27.4*	*29.2*	*34.2*	*1.3*	*6.1*	*442*	*12.7*	*88.7*	*21.5*	*1.20*	*1.08*	*1.36*	*45.5*	*55.9*
			mn	*16.9*	*19.3*	*14.0*	*24.0*	*0.88*	-	*291*	*3.6*	*66.1*	*21.0*	*0.74*	*0.53*	*0.67*	*37.1*	*45.7*
			mx	*34.2*	*50.5*	*60.9*	*56.6*	*2.3*	-	*560*	*27.6*	*136*	*22.0*	*1.77*	*1.66*	*2.47*	*67.3*	*82.8*

Table 3. *Generalized data for some parameters depending on soil type (designations are in the text)*

#	Tr	ST	Ra_{eq}			$^{238}U/^{232}Th$			$^{226}Ra/^{238}U$			$^{210}Pb/^{226}Ra$			D, nGy/h			AEDE, μSv/y		
			av	*mn*	*mx*	*av*	*mn*	*mx*	*av*	*mn*	*mx*	*av*	*mn*	*mx*	*av*	*mn*	*mx*	*av*	*mn*	*mx*
1	I	Al-Cr	93.5	89.5	98.2	1.23	0.74	1.55	1.20	0.99	1.57	0.97	0.82	1.22	48.4	47.2	49.1	59.5	58.1	60.4
2		Cn-Cr	88.0	66.1	136	1.44	0.98	1.77	0.99	0.53	1.42	1.17	0.67	1.94	45.2	37.1	67.3	55.6	45.7	82.8
3		Cn	75.2	71.9	78.4	1.18	1.12	1.24	1.08	0.84	1.32	1.56	1.42	1.71	40.1	38.1	42.1	49.3	46.8	51.7
4		GC	79.5	-	-	1.32	-	-	1.66	-	-	0.98	-	-	37.8	-	-	46.5	-	-
5	II	Al-Cr	95.1	77.2	111	0.84	0.75	0.95	0.94	0.86	1.10	1.82	1.12	2.47	48.2	39.8	55.6	59.3	48.9	68.4

Discussion

All identified radionuclides, with exception of 137Cs, are naturally occurring. They are characteristic for the region of East Georgia, in particular, for soil in the strip along the river Mtkvari [13].

The content and concentration of naturally occurring radionuclides identified in the samples in general corresponded to those usually observed in various soils [2]. The high activity recorded in sample Sl-3 might reflect the influence of shallow-lying rocks at the locations. The possible migration of their microparticles and mixing with the soil layer could cause hyperactivity in soil collected from this site.

The observed peculiarities of the activity ratios, in particular, the dominance of heightened values of 238U/232Th ratio may be connected with various geochemical processes. So, for example, in works [2,3] it is stated that behavior of Th and U isotopes in aqueous medium considerable differs from each other because of their different solubility, that can cause in some cases appreciable deviations of their ratio from the average value. As noted above the investigated region is characterized by a complex hydrogeological structure. During the circulation of underground waters dissolution of uranium in deep-lying rocks and its carryover into the soil formations located above could cause their enrichment by 238U. The similar picture can take place for Ra too, which is easily leached and washed away by water. This circumstance in view of the other geochemical factors can lead to appreciable variations of the 226Ra/238U ratio causing deviations to a greater or lesser extent from the average and equilibrium values.

The observed differences in concentrations of radionuclide families in samples of various soils in territory I could be connected with the formation conditions of the genetic soil types. The difference in 238U/232Th ratio (and also, to a certain extent, the 226Ra/238U ratio) in territories I and II, appears to be connected with the hydrological processes.

A prominent feature recorded in the majority of samples was the high 210Pb/226Ra ratio – in marked excess of the equilibrium activity value. In previous studies this has been connected with excess 210Pb precipitation from the atmosphere. However, given the complex geological-tectonic structure of the investigated region it is possible that the release of radioactive radon gas from deep layers of rocks and soil could be the cause. This process can lead to the occurrence of a “nonequilibrium” concentration of 210Pb (so-called “allochthonous” 210Pb [14].

Radionuclide 7Be, 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 that could have led to a reduction in concentration to values below MA.

The distribution of radionuclide 40K was similar to values observed in the work [13].

Data for the technogenic radionuclide 137Cs are of special interest. Usually its presence is connected with the Chernobyl accident (1986). For the last period the activity of 137Cs has considerably decreased, due to washing away processes or migration into deeper layers that can provoke the big variations of its activity in samples collected in different locations.

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 (29-56 μSv/y) was below limiting value of 1 mSv/y [15-17].

Finally, the results represent doubtless scientific and applied interest in the investigated region, confirming the need for such studies, which should be carried out on a regular basis.

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References

[1] United Nations Scientific Committee on the Effects of Atomic Radiation. Exposures from natural sources of radiation. Report to the General Assembly, UNSCEAR 1993.

[2] R.Kogan, I. Nazarov, Sh. Fridman., Basics of Environmental Gamma-spectrometry, Atomizdat, Moscow, 1976 (in Russian).

[3] N.Titaeva, Nuclear Geochemistry, MGU, Moscow, 2000 (in Russian).

[4] G.Faure, Principles of Isotope Geology, Wiley, New York, 1986.

[5] N.A. Alnassar, M.S. Jaafar, N.A. Kabir, Determination of concentrations of natural radionuclides in soils and water in non-cultivated sites in Seberang Perai, Malaysia, IOSR J. of Appl. Phys., 9, 2 (2017) 27-35.

[6] R. Mehra, M. Singh, Measurement of radioactivity of 238U, 226Ra, 232Th and 40K in soil of different geological origins in Northern India, J. of Envir. Prot., 2 (2011) 960-966.

[7] A. Klos, Z. Ziembik, M. Rajfur, A. Dołhańczuk-Śródka, Z. Bochenek, J. W. Bjerke, H. Tømmervik, B. Zagajewski, D.Ziólkowski, D. Jerz, M. Zielińska, P. Krems, P. Godyń, The origin of heavy metals and radionuclides accumulated in the soil and biota samples collected in Svalbard, near Longyearbyen, Ecol.Chem. Eng.S., 24(2) (2017) 223-238.

[8] N. Pallavicini, Activity concentration and transfer factors of natural and artificial radionuclides in the Swedish counties of Uppsala and Jämtland, Master's Thesis in Environmental Science, Swedish University of Agricultural Sciences, Uppsala, 2011.

[9] E. Charro, A. Moyano, Soil and vegetation influence in plants natural radionuclides uptake at a uranium mining site, Radiat.Phys. and Chem., 141 (2017) 200–206.

[10] L. Mtsariashvili, N. Kekelidze, B. Tutberidze, E. Tulashvili, I. Ambokadze, Radon in natural waters in Tbilisi artesian basin, Georgia, Proc. of 16th Int. Multidisciplinary Scientific GeoConference; Bulgaria; Book 3, 1 (2016) 605-612.

[11] 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.

[12] United Nations Scientific Committee on the Effect of Atomic Radiation. Exposure from natural radiation sources. Report to the General Assembly, UNSCEAR 2000.

[13] NATO-OSCE Project SfP 977991 "South Caucasus River Monitoring", NATO Science for Peace Programme. 2002-2008. www.kura-araks-natosfp.org

[14] T. Goslar, M.. Ganowicz, J. Czernik, and J. Sikorski, First measurements of natural radioactivities of 210Pb in the Institute of Physics, Silesian University of Technology. Geochronometria, 18 (2000) 29-34.

[15] Lj. Janković Mandić, S. Dragović, Assessment of terrestrial gamma exposure to the population of Belgrade (Serbia), Radiat.Prot.Dosim, 140, 4 (2010) 369–377.

[16] United Nations Scientific Committee on the Effect of Atomic Radiation. Sources, effects and risks of ionizing radiation. Report to the General Assembly, UNSCEAR 1988.

[17] International Commission on Radiological Protection. The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103. Ann. ICRP 37, Oxford: Pergamon, 2007, pp. 80-92.