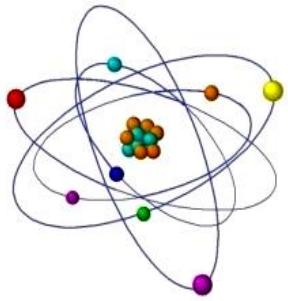
GAMMA-SPECTROSCOPY MEASUREMENTS OF RADIOACTIVITY AND ASSESSMENT OF RADIATION HAZARD INDICES IN SOILSAMPLES FROM SOME REGIONS OF THE BLACK SEA COAST



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ABSTRACT: *There are given results of radioactivity research in soil samples within the territory of some regions of the Black Sea coast. Ten samples have been collected from 10 locations. Up to 22 radionuclides were identified. Activity concentration of radionuclides of Th-232 family was in limits from 15.2 to 32.5Bq/kg (mean, 24.5Bq/kg), U-238 family – from 9.7 to 23.8 Bq/kg (mean, 17.2Bq/kg), U-235 family - from 0.62 to 1.31Bq/kg (mean, 0.93Bq/kg). Also individual radionuclides have been identified: K-40 – from 81.1 to 485Bq/kg (mean, 300Bq/kg), Cs-137 – from 8.8 to 191 Bq/kg (mean, 68.4Bq/kg); Be-7 – from 2.2 to 29.5 Bq/kg (mean, 12.3 Bq/kg). Radium equivalent activity varied from 44.9 to 89.4Bq/kg (mean, 73.2Bq/kg). Annual effective dose varied from 0.027 to 0.054mSv/y (mean, 0.043 mSv/y).*

Key words: radionuclides, soil, activity concentration, Black Sea coast

INTRODUCTION

Soil radioactivity is one of the main components of the Earth's radioactive background. Radioactivity in various areas of the globe differs considerably – from units up to hundreds of Bq/kg. Therefore, studying the terrestrial radioactivity in each territorial region is very important. Such studies are carried out in many countries. So, for example, in the work [1] 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 – CalcaricFluvisols, EutricGleysols, CalcaricRegosols, RendsicLeptosols, HaplicKastanozems, and HaplicPhaeozems; it was determined activity concentration of Th-232, U-238, Ra-226, Pb-210, Pb-210ex, K-40, Cs-137; the correlation between soil radioactivity and its mineralogical content was investigated. In another work [2] the activity of natural radionuclides in samples from 10 different regions in Qena governorate and Wadi EL-Lagita (Egypt) was investigated. Concentrations of radionuclides in soils ranged from 7.9±2.8 to 96.1±9.8 for Ra-226, 8±2.8 to 19 ±4.4 for Th-232 and 85.2±9.2 to 302.5±17.4 Bq/kg for K-40. The radiological health implication to the population that may result from these values is found to be low and almost insignificant, except in one case. No artificial radionuclide, however, was detected in any of the samples; hence, measurements have been taken as representing baseline values of these radionuclides in the soil in the studying areas. Similar research on naturally occurring radioactive materials (NORM) in the soil of Yalova, northwestern Turkey has been carried out in the work [3]. In addition, maps for the radionuclide activity concentrations of soil and the outdoor gamma dose rate distributions have been plotted for the region. The results of the study were discussed with the studies done in the close cities and the worldwide averages. In another work [4] soil samples (together with cement and fertilizers samples) collected in Sergipe State (Brazil) were studied; the radionuclide activities are below the Brazilian limit of the

exclusion and exemption criteria from the requirement of radiation protection. It also detected an unexpected Cs-137 in some samples; however, its activities do not represent a risk for the population. Surface and depth profile concentrations (down to 50 cm) of Th-232Th chain, Ra-226, and K-40 radionuclides were determined in undisturbed coastal and inland soils of La Plata city region, Argentina [5]. No dependence of the activity of the Th-232 natural chain on depth was found, whereas variations for Ra-226 and K-40 activities were observed. Positive correlations, determined by the Pearson correlation coefficients, were established between K-40, Ra-226 and Th-232 activity concentrations for the whole set of soil 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 [6]. In Georgia, regular research of natural (and also technogenic) radioactivity was not actually carried out. Rather detailed research of radioactivity in various environmental objects have been carried out in 1986, after the failure of the Chernobyl atomic power station and, basically, concerned technogenic radionuclides [7, 8, 9]. In these works it has been shown, that during this period in the territory of Western Georgia, basically in the strip adjoining the sea big concentrations of technogenic radionuclides were observed (in particular, Cs-137 concentration made from several hundred to some thousands of Bq/kg). It is possible to note also works [10, 11] in which results of research of radiation condition of the coast of the water area of the Black sea during a 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 the 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 [12]. Urushadze and Manakhov studied the content of technogenic radionuclides Cs-137 and Sr-90 in different types of soil in the territory of Georgia [13].

In the present work, there are given results of radioactivity research of soil samples within the territory of several regions of the Black Sea coast, Georgia.

MATERIALS AND METHODS

Study area

The study area of the Black Sea coast settles down between the settlement of Gonio and the city of Poti. In this area soil of two types – alluvial and red soils are most extended. Samples were collected near to following settlements and cities – Gonio, Batumi, Chakvi, Kobuleti, Shekvetili, Grigoleti, and Poti. In total 10 soil samples have been collected. The list of the investigated samples and their types is given in Table 1.

Table 1

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

#	CP	Lt(N); Ln(E)	ST
1	CP-1	41.554898; 41.578068	Rd
2	CP-2	41.605098; 41.578903	Al (Ac)
3	CP-3	41.616148; 41.587689	-“-
4	CP-4	41.636601; 41.655869	Al (St)
5	CP-5	41.669682; 41.691541	-“-
6	CP-6	41.733941; 41.738102	Rd
7	CP-7	41.804014; 41.777397	Al (St)
8	CP-8	41.958202; 41.762573	-“-
9	CP-9	42.018461; 41.762107	-“-
10	CP-10	42.184805; 41.648328	-“-

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

Sampling and analysis

Sample collection and preparation

Representative soil samples were collected from the soil surface (depth, 0.30 m) with the use of the special hand auger directly in plastic containers (volume up to 2.0 L). After drying in laboratory conditions samples were ground and sieved for their homogenization. Then samples were dried at the temperature of 105 - 110°C to constant weight and their bulk density and weight were determined. These values were used in the description of sample geometry. The samples were tightly sealed in Marinellibeaker (besides polyvinyl chloride adhesive tape was used also for hermetic sealing) and stored for 4 weeks before gamma-spectrometric analysis to avoid the escape of Rn-222 gas to attain a secular equilibrium between Ra-226, Th-232 and their respective progenies.

Gamma spectrometric analysis

The samples were analyzed for their radionuclides contents and activity concentrations 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 concentration of Th-232 was determined as averaged value for Ac-228, Ra-224, Pb-212, and Bi-212 (which the determination error varied from 1.4% to 6.4%), and that of U-238 was determined as averaged value for Th-234, Pb-214, Bi-214 (which the determination error varied from 7.1% to 16.2%). Since K-40 is directly a gamma-emitter, its activity concentration could be determined from its single photopeak at 1460 kev. Also identified was the technogenic radionuclide Cs-137. In samples "super-equilibrium" (allochthonous) Pb-210 (Pbal) was observed, the value of which was determined as the difference between measured activity values of Pb-210 and Ra-226 [1].

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 [14]:

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

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

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

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 - 0.2, D – absorbed dose rate *D* (nGy/h):

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

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

In the case of the presence of allochthonousPb, the contribution of its "allochthonous" parents Pb-214 and Bi-214 (and, accordingly, the activity *Raeq-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 [15], 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 • *ARa-al was* added to the calculation formula (1) where *ARa-al* is equal to *A*Pb-al = *A*Pb - *A*Ra [1]. A similar term (0.456•*ARa-al*) was added into equation (3) for the absorbed dose rate *Dal* and, accordingly, was considered for the calculation of AEDE*al*.

For samples characterization by the degree of radioactivity taking into account the accepted limiting

value *Raeq*- 370 Bk/kg (equivalent to γ-radiation dose of 1.5 mSv/y) [16] there were established several groups of samples by the value of radium equivalent activity, in particular: 1st group - not radioactive samples with activity no more than 30 Bq/kg; 2nd group – with low radioactivity in the range from 30 to 100 Bq/kg; 3rd group – with average radioactivity in the range from 100 to 300 Bq/kg; 4th group – samples with the raised radioactivity in the range from 300 to 1000 Bq/kg. The technique is described in more detail in works [17, 18].

RESULTS AND DISCUSSION

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.

The activity of identified radionuclides of the different families varied from 0.62 Bq/kg (for the U-235 family) to 65.5 Bq/kg (for the U-238 family). Among individual radionuclides, K-40 had the greatest activity (up to 485 Bq/kg). The activity of several radionuclides in some samples was below the minimum detectable activity (MDA). Activity concentrations of the main radionuclides in the studied samples, radium equivalent activity with no account taken of allochthonous Pb-210al (Raeq) and taking this into account (Raeq-al) and, accordingly, annual effective dose (AEDE and AEDEal), their average (*av*), minimal (*mn*) and maximal (*mx*) values are given in Table 2.

Table 2

The activity concentrations (A, Bq/kg) of the radionuclides, radium equivalent activity with no account taken of allochthonous Pb-210_{al} (Ra_{eq}) and taking this into account ($Ra_{eq\text{-}al}$) and, accordingly, annual effective dose (AEDE and $AEDE_{al}$), their average (*av*), minimal (*mn*) and maximal (*mx*) values

CP	A, Bq/kg											Ra_{eq}	$Ra_{eq\text{-}al}$	AEDE	$AEDE_{al}$
	Th-232	U-238	Ra-226	Pb-214	Bi-214	Pb-210	U-235	Be-7	K-40	Cs-137	Pb-210_{al}	Bq/kg	Bq/kg	mSv/y	mSv/y
CP-1	26.3	18.8	28.8	16.9	17.0	53.6	1.04	6.7	471	83.7	24.8	89.4	114	0.054	0.068
CP-2	15.2	9.7	14.0	8.0	7.7	43.6	0.62	4.9	192	21.1	29.6	44.9	74.1	0.027	0.043
CP-3	20.8	13.6	20.0	10.9	10.7	56.7	0.89	23.5	259	95.1	36.7	61.5	97.6	0.036	0.057
CP-4	32.5	23.8	34.8	21.6	21.5	55.2	1.31	29.5	195	47.6	20.4	83.9	104	0.048	0.059
CP-5	24.3	17.3	28.7	17.6	17.0	27.1	0.80	10.1	328	56.9	-1.6	75.0	73.5	0.045	0.044
CP-6	23.5	16.2	19.7	12.6	12.2	51.1	1.08	16.3	81	66.0	31.4	55.4	86.4	0.031	0.048
CP-7	18.7	16.3	16.3	14.4	13.7	65.5	0.92	5.4	485	191	49.1	77.0	125	0.048	0.076
CP-8	28.4	20.5	31.7	19.6	19.3	41.1	1.05	-	261	52.2	9.4	79.5	88.7	0.046	0.051
CP-9	32.4	17.9	22.3	18.5	18.4	17.4	0.80	2.2	292	8.8	-4.8	84.6	79.9	0.049	0.046
CP-10	22.6	17.6	22.1	18.5	17.1	33.6	0.81	-	436	61.4	11.4	80.5	91.8	0.049	0.056
av	24.5	17.2	23.8	15.9	15.5	44.5	0.93	12.3	300	68.4	20.6	73.2	93.5	0.043	0.055
mn	15.2	9.7	14.0	8.0	7.7	17.4	0.62	2.2	81.1	8.8	-4.8	44.9	73.5	0.027	0.043
mx	32.5	23.8	34.8	21.6	21.5	65.5	1.31	29.5	485	191	49.1	89.4	125	0.054	0.076

Within families of radionuclides, activity varied within sufficiently wide limits. In particular, the range (mean) was 5.5-35.0 (20.9) Bq/kg for Th-232, 7.7-65.5 (23.8) Bq/kg for U-238, and 0.62-1.31 (0.92) Bq/kg for U-235.

The activity of K-40 varied from 81.1 to 485 Bq/kg (mean, 300).

The mean activity concentration of Th-232 estimated in this study is 20.9 Bq/kg, which is lower than the world average of 33Bq/kg. The mean activity concentration of U-238 estimated in this study is 23.8

Bq/kg, and is thus lower than the world average of 45 Bq/kg. The main activity concentration of K-40 estimated in this study is 300 Bq/kg, which is lower than the world average of 400 Bq/kg.

Figure 1 shows the statistically significant correlation (R = 0.7272) between the Th-232 and U-238 activity concentrations.

Radionuclide Be-7 was observed in actually all samples with activity in the range 2.2-29.5 Bq/kg. Technogenic radionuclide Cs-137 was measured in all samples and was found to be in the range of 8.8 to 191 Bq/kg (mean, 68.4).

All samples by the level of equivalent activity belonged to the group with low radioactivity.

The highest values of equivalent activity were observed for soil type Al(St), with an average value of 80.1 Bq/kg, and rather less for soils Rd – 72.4 Bq/kg, and the least value was observed for Al(Ac) – 53.2 Bq/kg.

Determined minimal and maximal values of annual effective dose varied in the range of 0.027-0.054 mSv/y. These values (as well as equivalent activity) increase (annual effective dose - in the range 0.043-0.076 mSv/y) under the assumption, that allochthonous Pb-210 is caused by excess soil radon.

It is common knowledge that the concentration of radioactive elements in soils is determined by the radioactivity of initial rocks and the subsequent soil formation processes. The content and concentration of naturally occurring radionuclides, in general, correspond to those usually observed for different soils [19].

Due to specific processes of soil 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 the results, it is not observed the noticeable expressed dependence on soil type.

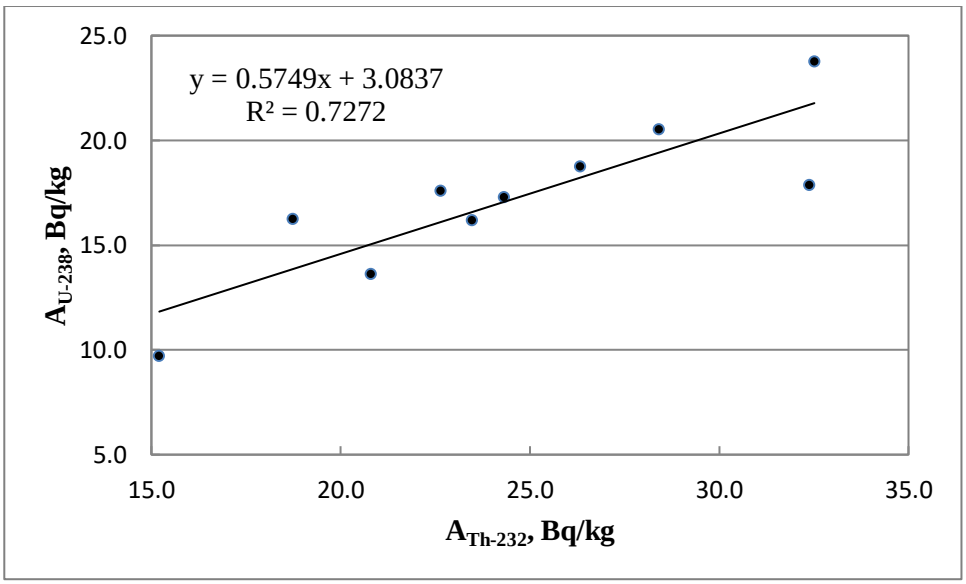

Figure 1. Correlation between Th-232 and U-238 activity concentration.

The presence of “super equilibrium” (allochthonous) Pb-210 connects with radon migration from the bottom layers of the earth to the top (and further in the atmosphere). Pb-210 is capable to accumulate on the walls of pores and faults at the passage of radon flow through the 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 the soil is thus broken towards an increase in activity of Pb-210. In a number of works presence of “super equilibrium” Pb-210 in the soil is connected with its deposition from atmospheric air.

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

Technogenic radionuclide Cs-137 was observed in all samples in sufficiently appreciable amounts. Usually, its occurrence is connected with the 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 [20], values of Cs-137 activity are now, basically, in the range of 1 - 10 Bq/kg. With a certain degree of convention, it is possible to consider this level as background value for the whole territory of Georgia. The average value (68.4 Bq/kg) is greater than this quantity which can be due to non-uniform precipitations following the accident. The distribution of the naturally occurring radionuclide K-40 was similar to values observed by Kogan et al. [19].

The calculated values of the annual effective dose do not exceed the 1 mSv/y dose limit recommended for public radiation exposure control [21,22,23]. Table 3 compares the activity concentrations of radionuclides in investigated samples in the present study with those by other investigators in various countries of the world. 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 the urgency of such research and the necessity of their regular character.

Table 3

Activity concentration (A, Bq/kg) of radionuclides and some other parameters in soil in different regions of the world

SR	A, Bq/kg					Ra_{eq}, Bq/kg	AEDE, mSv/y	Ref
	Th-232	U-238	Ra-226	K-40	Cs-137			
Sp	34.6	40.2	26.7	586.2	30.9			[1]
	23.7-49.4	19.9-60.0	20.8-34.9	446-799	4.4-64.7			
It	48	79		640	25.0			[24]
	20-70	24-231		242-1434	1.1-241			
Tk	51.8	24.5		344.9	26.3	125.0	0.070	[25]
	9.5-170.8	7.4-79.8		35.7-913.8	0.6-154.3	23.8-293.6	0.013-0.192	
Ch	101.0	79.3	75.1	535.8		260.8	0.147	[26]
	10.3-376.0	12.0-264.3	7.6-298.3	8.2-1747.1		28.3-850.3	0.017-0.468	
In	64.5		60.3	481.0		189.5	0.37	[27]
	36.1-136.1		15.0-205.6	65.8-795.9		110.0-436.2	0.06-0.24	
Ng	85.84		52.05	477.69	1.60	210.57	0.1170	[28]
	13.54-295.24		8.33-160.37	15.97-2723.22	0.89-3.53	28.47-701.53	0.0152-0.3891	
Ge	24.5	17.2	23.8	300	68.4	73.2	0.043	Present
	15.2-32.5	9.7-23.8	14.0-34.8	81-485	8.8-191	44.9-89.4	0.027-0.054	study
Ww	45	33	32	412				[29]
	0.05-360		0.5-1000	4-3200				

Note. Studied regions (SR): Sp – Spain; It – Italy; Tk – Turkey; Ch – China; In – India; Ng – Nigeria; Ge – Georgia; Ww - Worldwide average values.

CONCLUSION

Up to 22 radionuclides are identified in soil samples selected from the ten locations in some regions on the Black Sea coast. The mean activity concentrations of naturally occurring radionuclides Th-232 and U-238 families were lower than the world average values as well as the mean activity concentration of K-40. Technogenic radionuclide Cs-137 was measured in all samples and was found to be in the range of 8.8 to 191 Bq/kg (mean, 68.4). Comparison with reference data as well as analysis of obtained results was carried out.

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