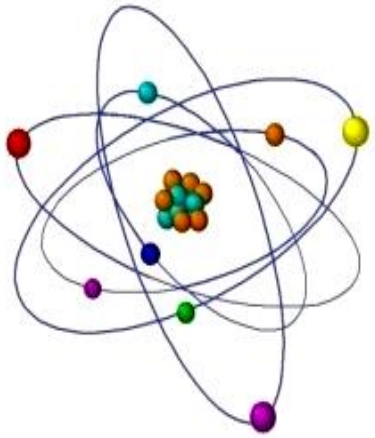
NATURAL RADIOACTIVITY CONTENT OF THE LIMESTONE AND SANDSTONE ROCK SAMPLES IN THE CHIATURA REGION

(SOUTH CAUCASUS, GEORGIA)



Eremia Tulashvili*, Mariam Akhalkatsishvili, Lela Mtsariashvili, Manana Chkhaidze

Iv. Javakhishvili Tbilisi State University,
Faculty of Exact and Natural Sciences,
N. Kekelidze Material Research Institute, Georgia

DOI:10.63465/rrs6202611756 *Corresponding author: eremia.tulashvili@tsu.ge

ABSTRACT: *There are presented the results of the first conducted research of the natural gamma-radioactivity of limestone and sandstone rock samples from the Chiatura region, West Georgia. The study was carried out using a Canberra GC2020 germanium gamma-spectrometer. Twenty-one natural and one technogenic gamma-emitted radionuclides were identified in the studied samples. The activity concentration of the identified radionuclides was determined. The radium equivalent activity ranges from 19.2 Bq/kg to 91.7 Bq/kg with an average of 37.0 Bq/kg for limestone samples and from 32.9 Bq/kg to 374 Bq/kg with an average of 133 Bq/kg for sandstone samples. The absorbed dose rate ranges from 8.96 nGy/h to 42.6 nGy/h (an average of 17.3 nGy/h) for limestone samples, and from 16.0 nGy/h to 176 nGy/h (and average of 64.4 nGy.h) for sandstone samples. There were calculated also the external hazard index and the internal hazard index. The data obtained were compared with literature data. The hazard indices were found to be within safety limits.*

Key words: Radioactivity, Rocks, Radiological indices

INTRODUCTION

Radioactivity is a part of the natural environment: the Earth's surface, rocks, atmosphere, human bodies, and food. The majority of the Earth's radioactive elements are found in the rocks that make up the Earth's crust. From there, radioactive elements are transferred to the soil, then to plants, and finally to the bodies of animals and humans through plants. The radioactivity of rocks and ores is determined by the content of radioactive rock-forming minerals. Depending on the qualitative and quantitative composition of these minerals (in particular, uranium-containing rocks), the conditions of formation, age, and degree of metamorphism, their radioactivity varies greatly. The radioactivity of rocks and ores is higher the higher the concentration of minerals containing natural radioactive elements, mainly from the uranium and thorium families, and potassium-40.

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

For example, samples of sedimentary (shales, limestones, and sandstones) and metamorphic (black and asphalt shales, graphite, marble, quartz-mica, calcareous schist, and quartz) rocks from the state of Kashmir (Pakistan) were studied in the work [1]. In the samples of sedimentary rocks, the activity concentration of Ra-226 varied from 18.1 to 45.2 Bq/kg (average value of 31.4 Bq/kg), Th-232 varied from 28.7 to 68.9 Bq/kg (average value of 48.5 Bq/kg), and K-40 varied from 224 to 610 Bq/kg (average value of 381 Bq/kg). The authors noted that the Cs-137 activity concentration values in all samples were below the detection limit. The radium equivalent activity of the samples was estimated, which ranged from 20.6 to 294 Bq/kg (with an average activity value of 126 Bq/kg), which is significantly lower than the recommended limit of 370 Bq/kg [2], as recommended by [3], for the external dosage to be less than 1 mSv/y.

In another works [4] samples of Tufa deposits from four separate occurrences in the Dinaric Karst of Croatia was examined. It was found that the concentration of Th-232 activity varied in the range of 1.0 - 12.9 Bq/kg with an average value of 4.5 Bq/kg; the average concentration of U-238 was noticeably higher and amounted to 7.5 Bq/kg (range 1.3 - 14.6 Bq/kg); results comparable to U-238 were obtained for Ra-226 is the average value of 7.3 Bq/kg, the range is from 2.9 to 13.0 Bq/kg. The studied samples also contained the technogenic radionuclide Cs-137, with an activity concentration ranging from 3.9 to 77.8 Bq/kg, with an average value of 24.6 Bq/kg. The authors attribute this to Cs-137 contamination from rainwater, as the outdated limestone tuff is not a closed system, and the sampling was conducted after the 1986 accident, which was accompanied by heavy rainfall in the area.

The ratios of radionuclide activities were also calculated, with values for U-238/Th-232 ranging from 0.81 to 5.30, with an average value of 2.41, and for Ra-226/U-238 ranging from 0.59 to 2.22, with an average value of 1.10.

Various rocks used as building materials in Romania were studied [5]. Among them was tuff, for which the activity concentrations of Th-232, Ra-226, K-40, and Cs-137 were 9.8, 10.5, 180, and 9.6 Bq/kg, respectively.

Some tens of limestone samples collected from all over Turkey were studied in the work [6]. The activity concentration of Ra-226 varied from 0.7 to 55.1 Bq/kg with an average value of 19 Bq/kg, Th-232 varied from 1.2 to 20.9 Bq/kg with an average value of 4.3 Bq/kg, and K-40 varied from 10.1 to 258.4 Bq/kg with an average value of 55 Bq/kg. The calculated values of equivalent activity varied within the interval 3.2–81.6 Bq/kg, with an average value of 29.4 Bq/kg. Research on the radioactivity of various environmental objects in Georgia has been carried out in the past, stimulated by the disaster at the Chernobyl atomic power station in 1986. Research is being conducted on the natural radioactivity of various environmental structures [7, 8]. The present work analyzes the natural (and technogenic too) gamma radioactivity of sedimentary rocks, in particular limestone and sandstone rocks in the Chiatura region (South Caucasus, West Georgia), which are widely used as raw materials for construction purposes.

The aim of the study was to determine the radionuclide composition and background concentrations of radionuclides in samples of rocks in the study area, to study the level of gamma radioactivity, and to assess the corresponding radiological risk for the local population. This region is one of the most important industrial zones of Georgia, as it is home to one of the world's largest manganese deposits. The Chiatura deposit has large reserves of high-quality manganese ores, and from the beginning of its operation (1879) it has occupied a prominent place in the world. The study of the radioecological condition of the territory of this region has not been conducted before, which necessitated the implementation of this work.

MATERIAL AND METHODS

Study area. The study area, which is a high plateau (at an altitude of 340-500 meters above sea level), is located within the Dzirula crystalline massif (in its northwestern part). This area is divided into separate highlands by the Kvirila River and its tributaries. The area is mainly composed of Mesozoic and Paleogene-Neogene sedimentary rocks. Spongolite sandstones are found in the western part, while the eastern and north-eastern parts are predominantly composed of thin-layered clay rocks.
Control points and samples. In the study area 16 limestone and 17 sandstone (of different compositions) rock samples have been collected from 31 control points in the city of Chiatura and nearby villages – Perevi, Shukruti, Ithvisi, Tabagrebi, Mgvimevi, Darkveti, Rgani (three samples were selected at control point Ch-35). Table 1 lists the control points, their geographical coordinates, and the types of samples selected.
Sample collection and preparation. Samples were selected from outcropped rocks and put into plastic containers (volume up to 2.0 liters). After drying in the laboratory, samples were broken into pieces < 40 mm and were then crushed using a special BB 50 jaw crusher (Retcsh GmbH, Germany) to a size of approximately 1 mm. Samples were then dried at 105°C to constant weight, and their bulk density was determined. These values were used for the description of sample geometry. The dried samples have been filled in a 1 L Marinelli beaker of polypropylene and stored for more than 4 weeks to achieve secular equilibrium between radium and its decay products.

Measurements

Gamma radiation activity

Measurements were carried out using a gamma spectrometer Canberra GC2020 with a semiconductor germanium detector with a relative efficiency of 24%. The gamma spectra acquisition time was 48 hours. Genie-2000 S500 software with additional modules was used for the analysis, in particular, the S506 an Interactive Fit Program. With this program, “decomposition” of the interference peak in the area of 186 keV was carried for all spectra (the program identifies one peak in this area that emerges from the interference of two closely spaced peaks U-235 (185.715 keV) and Ra-226 (186.211 keV)). The S506C program processes the spectral curve mathematically, meaning that two Gaussian peaks are created in this area with energies corresponding to U-235 and Ra-226. In the identification of peaks and the calculation of activity concentration, a tolerance value was established in such a manner that low-energy peak was compared only with U-235 and the high-energy peak only with Ra-226. The results show that the uncertainty in the determination of the activity concentration of Ra-226 was between 8.8 and 24%. Its activity was compared with the activity of its daughters, Pb–214 and Bi-214, which had an uncertainty between 2.4 and 3.0%.

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

#	CP	Lt(N); Ln(E)	SN	ST	#	CP	Lt(N); Ln(E)	SN	ST
1	Ch-1	42.2449; 43.3039	442	Ls	18	Ch-19	42.3250; 43.3261	475	Ss
2	Ch-2	42.2865; 43.3099	444	Ls	19	Ch-20	42.3222; 43.3420	477	Ss
3	Ch-4	42.2995; 43.3245	447	Ls	20	Ch-21	42.3062; 43.3283	478	Ss
4	Ch-5	42.3156; 43.3394	450	Ls	21	Ch-22	42.3048; 43.3266	480	Ss
5	Ch-6	42.3084; 43.3294	451	Ls	22	Ch-23	42.3023; 43.2553	482	Ls
6	Ch-7	42.2955; 43.2975	453	Ls	23	Ch-24	42.3010; 43.2615	485	Ss
7	Ch-8	42.2812; 43.2845	454	Ls	24	Ch-25	42.2992; 43.2634	487	Ss
8	Ch-9	42.3055; 43.3008	455	Ss	25	Ch-26	42.2978; 43.2643	488	Ss
9	Ch-10	42.3057; 43.3009	457	Ss	26	Ch-27	42.2914; 43.2917	490	Ss
10	Ch-11	42.3038; 43.3143	459	Ls	27	Ch-29	42.3114; 43.2693	492	Ss
11	Ch-12	42.3029; 43.3143	461	Ls	28	Ch-30	42.3099; 43.2723	495	Ls
12	Ch-13	42.3024; 43.3138	463	Ls	29	Ch-35	42.3148; 43.2674	498	Ss
13	Ch-14	42.3020; 43.3130	465	Ls	30	"-"	"-"	499	Ss
14	Ch-15	42.3198; 43.2884	467	Ls	31	"-"	"-"	501	Ls
15	Ch-16	42.3082; 43.2904	469	Ls	32	Ch-37	42.3132; 43.3934	504	Ss
16	Ch-17	42.2907; 43.2918	472	Ss	33	Ch-38	42.3214; 43.3977	505	Ss
17	Ch-18	42.3259; 43.3270	474	Ss					

Notes: 1) Lt(N) – latitude (north); Ln(E) – longitude (east). 2) SN – sample number.
3) *Rock types*: Ls – Limestone; Ss - Sandstone;

The values for the activity of Ra-226, Pb-214 and Bi-214 did not differ significantly. It is therefore to consider that similar determination uncertainty of Ra-226 concentration is satisfactory; thus, this method was also used for the determination of U-235 activity concentration based on the 185.715 keV line. The observed values for U-235 activity were compared to the values for U-238 activity (which were determined based on the 63.3 keV line of Th-234, with an uncertainty range of 6.3 - 14%). The value of the activity ratio U-238/U-235, which is considered to be constant (21.7) for natural objects [9], was used as a criterion. With large deviations from this value (more than 10%), the 185.715 keV line

(as well as the 63.3 keV line) was reanalyzed using the S506 program. On occasion, in low-activity samples, the Ra-226 activity was specified by the average activity of Pb-214 and Bi-214, and the obtained value was considered to be a definitive estimation of U-235 activity. To determine the activity of Th-232, the average values for Ac-228, Ra-224, Pb-212, Bi-212 were used, the errors of which ranged from 3.2 to 7.2%.

Taking into account the influence of matrix composition, the chemical composition of samples was determined on the basis of data from the literature [10], which were then used in special software (LabSOCS) to calibrate the efficiency of the activity concentration. The LabSOCS system allows the user to create calibrations based on laboratory quality efficiency without using radioactive calibrated sources. For radionuclide identification, a special library was used containing the lines of 41 radionuclides and other specific sources (351 lines in total). The NuDat database [11] was used for library compilation.

Radiological indices

Assessment of radium equivalent activity Ra_{eq} (Bq/kg) and annual effective dose equivalent *AEDE* (mSv/y) was carried out under Eq. 1 and Eq. 2 [2]:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad (1)$$

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, 0.7×10^6 μSv/Gy, k_2 – outdoor occupancy factor (the fraction of time spent in the open air) which equals to 0.2, D – absorbed dose rate (nGy/h), which is calculated using Eq. 3:

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

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

Other indices were also calculated to assess the radiation hazard, including:

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

$$H_{in} = (A_U/185) + (A_{Th}/259) + (A_K/4810) \quad (5)$$

The values of the indices (H_{ex}, H_{in}) must be less than unity for the radiation hazard to be negligible [12].

RESULTS AND DISCUSSION

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 concentrations of the main radionuclides of the investigated samples, the equivalent activity, the activity ratios and their averages (*av*), the minimal (*mn*) and maximal (*mx*) values, among other data, are given in Table 2.

Activity concentration. The activity concentrations of natural radionuclides vary significantly in different samples, in particular, values of activity of radionuclides U-238, Th-232, U-235 and K-40 vary for limestone and sandstone samples, respectively, in the ranges 16 – 87.4 Bq/kg (average value of 32.8 Bq.kg), 0.43 – 2.86 Bq/kg (1.38 Bq/kg), 0.74 – 4.58 Bq/kg (1.65 Bq/kg), 11.7 – 91.8 Bq/kg (32.5 Bq/kg) and in the ranges 12.8 – 286 Bq/kg (76.0 Bq.kg), 6.62 – 33.9 Bq/kg (18.6 Bq/kg), 0.76 – 13.9 Bq/kg (3.80 Bq/kg), 133 – 762 Bq/kg (433 Bq/kg). As can be seen from Table 2, the average activity of radionuclides of the U-238 family for limestone samples (32.8 Bq/kg) is more than two twice lower that of sandstone (76.0 Bq/kg); limestone samples are also characterized by a noticeably lower average activity of Th-232 family radionuclides (1.38 Bq/kg) compared with sandstone samples (18.6 Bq/kg). The average activity of the radionuclide K-40 for limestone samples is significantly lower than that of sandstone samples (32.5 Bq/kg compared to 433 Bq/kg). The cosmogenic radionuclide Be-7 was detected for several samples, with an activity ranging from <M to 7.90 Bq/kg (in some samples, it was detected in trace amounts). The technogenic radionuclide Cs-137 was detected in 18 samples in small amounts (<M - 1.40 Bq/kg), and in some samples, it was detected in trace amounts.

Activities ratio U-238/U-235 was calculated, and the values ranged from 18.1 to 21.6 (an average value 20.1) and from 16.4 to 22.0 (19.6) for limestone and sandstone samples, respectively, which is generally consistent with the value of 21.7 accepted for natural objects. The U-238/U-235 ratio values obtained for all samples are within the error margin of the natural value, which, in addition to the methodological aspect, allows us to conclude that there is no anthropogenic U-235 contamination.

Table 2. Activity concentration (*A*, Bq/kg) of radionuclides, activity ratio U-238/U-235, radium equivalent activity (Ra_{eq}, Bq/kg), absorbed dose rate (*D*, nGy/h), annual effective dose (*AEDE*, mSv/y), external hazard index (H_{ex}), internal hazard index (H_{in}), their average (av), minimal (mn) and maximal (mx) values

#	SN	*A*, Bq/kg						U-238/ U-235	Ra_{eq}, Bq/kg	*D*, nGy/h	*AEDE*, mSv/y	H_{ex}	H_{in}
		U-238	Th-232	U-235	Be-7	K-40	Cs-137						
						Limestone rock							
1	442	27.5±5.99	2.79±0.25	1.48±0.10	2.45±0.23	91.8±2.88	<M	18.6	37.9	18.2	0.022	0.10	0.18
2	444	37.6±8.12	1.52±0.11	1.77±0.09	-	17.5±0.67	0.08±0.02	21.3	41.0	19.0	0.023	0.11	0.21
3	447	30.9±6.70	0.78±0.04	1.49±0.10	<M	22.2±0.82	0.13±0.02	20.8	33.6	15.7	0.019	0.09	0.17
4	450	19.5±4.28	0.78±0.08	0.90±0.10	<M	15.2±0.59	<M	21.6	21.7	10.1	0.012	0.06	0.11
5	451	29.9±6.48	0.43±0.05	1.49±0.13	<M	11.7±0.51	<M	20.1	31.3	14.6	0.018	0.08	0.17
6	453	33.5±7.28	2.86±0.15	1.62±0.13	<M	84.1±2.66	1.40±0.05	20.7	43.4	20.7	0.025	0.12	0.21
7	454	18.5±4.06	0.92±0.09	0.92±0.09	2.04±0.21	12.0±0.50	<M	20.1	20.7	9.62	0.012	0.06	0.11
8	459	51.2±12.9	1.28±0.06	2.79±0.10	2.27±0.33	21.5±0.80	-	18.4	54.5	25.3	0.031	0.15	0.29
9	461	26.2±5.69	0.93±0.10	1.36±0.07	<M	14.7±0.59	0.20±0.02	19.3	28.6	13.3	0.016	0.08	0.15
10	463	29.0±6.31	1.59±0.12	1.61±0.13	-	38.6±1.31	0.26±0.02	18.1	34.0	16.0	0.020	0.09	0.17
11	465	44.3±9.56	2.22±0.14	2.22±0.16	<M	67.8±2.17	-	20.0	52.3	24.7	0.030	0.14	0.26
12	467	87.4±18.8	1.23±0.09	4.58±0.24	-	36.4±1.39	0.24±0.03	19.1	91.7	42.6	0.052	0.25	0.48
13	469	27.8±6.05	1.51±0.11	1.36±0.12	7.90±0.64	34.7±1.18	-	20.5	32.4	15.2	0.019	0.09	0.16
14	482	16.0±3.56	1.30±0.10	0.74±0.10	<M	18.3±0.72	<M	21.6	19.2	8.96	0.011	0.05	0.10
15	495	24.5±5.35	1.17±0.14	1.14±0.05	<M	20.1±	<M	21.5	27.6	12.9	0.016	0.08	0.14
16	501	20.9±4.58	0.77±0.12	1.03±0.09	<M	13.6±	0.24±0.02	20.3	23.0	10.7	0.013	0.06	0.12
	av	*32.8*	*1.38*	*1.65*	*3.66*	*32.5*	*0.36*	*20.1*	*37.0*	*17.3*	*0.021*	*0.10*	*0.19*
	mn	*16.0*	*0.43*	*0.74*	*<M*	*11.7*	*<M*	*18.1*	*19.2*	*8.96*	*0.011*	*0.05*	*0.10*
	mx	*87.4*	*2.86*	*4.58*	*7.90*	*91.8*	*1.40*	*21.6*	*91.7*	*42.6*	*0.052*	*0.25*	*0.48*
						Sandstone rock							
17	455	62.2±13.4	18.3±0.90	3.19±0.25	<M	289±8.75	-	19.5	109	51.9	0.064	0.30	0.47
18	457	80.3±17.2	27.4±1.22	4.11±0.29	-	511±15.4	<M	19.5	155	74.9	0.092	0.43	0.65
19	472	23.7±5.25	17.4±0.76	1.10±0.11	<M	655±19.6	-	21.6	94.4	48.8	0.060	0.27	0.33

20	474	14.4±3.30	14.0±0.63	0.88±0.08	-	435±13.1	0.17±0.02	16.4	65.0	33.3	0.041	0.18	0.22
21	475	12.8±2.92	7.73±0.43	0.76±0.11	<M	192±5.91	-	16.8	37.3	18.6	0.023	0.10	0.14
22	477	14.1±3.13	6.62±0.37	0.85±0.09	-	133±4.08	-	16.5	32.9	16.0	0.020	0.09	0.13
23	478	18.6±4.19	17.5±0.75	0.94±0.10	-	699±20.9	-	19.9	92.6	48.3	0.059	0.26	0.31
24	480	18.5±4.20	18.9±0.81	0.84±0.13	-	762±22.8	-	22.0	98.9	51.7	0.063	0.28	0.33
25	485	92.8±19.9	19.7±0.92	4.61±0.24	<M	469±14.1	<M	20.1	154	74.3	0.091	0.42	0.68
26	487	28.9±6.36	23.3±0.98	1.47±0.13	-	468±14.1	-	19.6	95.0	46.9	0.058	0.27	0.34
27	488	83.2±17.9	19.7±1.01	4.74±0.18	-	352±10.7	<M	17.5	136	65.0	0.080	0.37	0.60
28	490	88.3±18.9	16.9±0.92	4.41±0.28	-	294±8.97	-	20.0	133	63.2	0.078	0.36	0.60
29	492	74.1±15.9	21.2±0.99	3.38±0.19	<M	384±11.5	-	22.0	131	63.1	0.077	0.36	0.56
30	498	235±50.0	20.0±1.35	11.7±0.54	<M	332±10.1	-	20.1	286	134	0.165	0.78	1.41
31	499	117±25.1	21.9±1.21	5.89±0.30	<M	282±8.62	-	19.9	168	79.2	0.097	0.46	0.78
32	504	286±60.9	33.9±1.22	13.9±0.53	-	563±16.9	-	20.6	374	176	0.216	1.02	1.79
33	505	42.0±9.0	11.0±0.53	1.93±0.15	<M	542±16.2	<M	21.7	96	48.6	0.060	0.27	0.38
	av	*76.0*	*18.6*	*3.80*	<M	*433*	*0.10*	*19.6*	*133*	*64.4*	*0.079*	*0.37*	*0.57*
	mn	*12.8*	*6.62*	*0.76*	<M	*133*	*<M*	*16.4*	*32.9*	*16.0*	*0.020*	*0.09*	*0.13*
	mx	*286*	*33.9*	*13.9*	<M	*762*	*0.17*	*22.0*	*374*	*176*	*0.216*	*1.02*	*1.79*

Radiological indices

The average value of the radium equivalent activity Ra_{eq} for the limestone samples is more than three and a half times less than that of sandstone samples (37.0 and 133 Bq/kg, respectively). It should be noted that the Ra_{eq} values for almost of the samples studied (with the exception of one sample) are less than the recommended value of the equivalent radium activity of 370 Bq/kg. The values of the absorbed dose rate *D* vary in the range 8.96 – 42.6 nGy/h (17.3 nGy/h) and 16.0 – 176 nGy/h (64.4 nGy/h) for limestone and sandstone samples, respectively. As you can see, the average dose value for the limestone samples is approximately 3.7 times lower than that of the sandstone samples. It should also be noted that the dose values for all limestone samples are lower than the global average value of 55 nGy/h, and for almost all sandstone samples (with the exception of two samples), the dose values are approximately equal to the global average or slightly higher than the global average.

A similar pattern is observed for the values of the annual effective dose: the *AEDE* values vary in the range of 0.011 – 0.052 mSv/y for limestone samples and in the range 0.020 – 0.216 mSv/y for sandstone samples, and at the same time the average dose value for the limestone samples is approximately 3.7 times lower than that of the sandstone samples. All the obtained *AEDE* values are significantly lower than the recommended level of 1 mSv/y.

The H_{ex} and H_{in} values for almost all samples are less than one; the H_{in} index value exceeds unity only for three samples. Table 3 shows some literature data on the radiological indexes of limestone and sandstone rock samples obtained in some regions of the world. As can be seen from the results, the average values obtained in this study are approximately comparable to these results and are less than the recommended or restrictive radiation safety limits.

Table 3. Comparison of various data reported by researches from other regions

Region	*A*, Bq/kg			Ra_{eq}, Bq/kg	*D*, nGy/h	*AEDE*, mSv/y	Ref.
	U-238	Th-232	K-40				
Limestone rock							
Italy, Sicily	32.8 31.5 - 33.9	8.2 7.5 - 8.8	96.2 89.8 – 103.8	-	42.8	-	[13]
Tanzania	6.4 - 25.2	1.3 - 15.2	13.2 - 80	21.4 – 34.3	19.2 – 29.0	0.13 – 0.14	[14]
Kenya, Kerio Valley	104.785	40.317	427.408	207.1	90.6	0.1	[16]
Nigeria, Ewekoro	121.30 18.09 - 239.50	112.25 8.33 - 360.01	158.47 11.28 - 735.26	299.96 58.9 - 758.8	135.29 28.8 - 330.9	0.166 0.035 - 0.406	[17]
Iraq	28.38 – 64.12	-	290.5 – 637.1	-	105.3– 223.1	-	[18]
Georgia, Chiatura region	32.8 16.0 - 87.4	1.38 0.43 - 2.86	32.5 11.7 - 91.8	37.0 19.2 - 91.7	17.3 8.96 - 42.6	0.021 0.011 - 0.052	Present work
Sandstone rock							
Italy, Sicily	11.3 10.1 - 12.6	26.9 24.5 - 28.6	922 856 – 1011	-	100	0.247	[13]
Tanzania	7.7 - 18.3	12.8 - 30.3	180.4 – 501	39.9 - 100	35.6 – 90.2	0.17 – 1.13	[14]
Egypt, Fileita area	32.0 4.9 - 67.9	29.6 11.8 - 60.5	132.6 31.3 - 438.2	-	38 18 - 74	0.19 0.09 – 0.36	[15]
Kenya, Kerio Valley	72.707	64.180	677.387	228.1	100.6	0.1	[16]
Georgia, Chiatura region	76.0 12.8 - 286	18.6 6.62 – 33.9	433 133 – 762	133 32.9 - 374	64.4 16.0 - 176	0.079 0.020 - 0.216	Present work

CONCLUSION

For the first time the radionuclide composition and activity concentration of gamma-emitting radionuclides in limestone and sandstone rocks samples collected in the Chiatura region of Georgia was analyzed. Up to 22 radionuclides are mainly detected in the selected rock samples, in particular, of natural families Th-232, U-238, U-235, other natural radionuclides Be-7, K-40, technogenic radionuclide Cs-137. The values of the activity concentration of the identified radionuclides have been calculated. It has been shown that the values of the radium equivalent activity for all the samples studied are less than the maximum permissible value of 370 Bq/kg. The values of some radiological indexes have been calculated, indicating that the level of radioactivity in the samples studied does not exceed the recommended values and that the rocks studied do not pose any danger to the population.

Acknowledgements

This work was supported by Shota Rustaveli National Science Foundation of Georgia (SRNSFG) [grant number FR-23-2436].

RFERENCES

[1] Rafique, M., Khan, A.R. Jabbar, A., Rahman, S.U., Kazmi, S.J. A., Nasir, T., Arshed, W., & Matiullah. (2014). Evaluation of radiation dose due to naturally occurring radionuclides in rock samples of different origins collected from Azad Kashmir. Russian Geology and Geophysics, 55, 1103–1112.

[2] UNSCEAR: Sources and effects of ionizing radiation: Exposure from natural radiation sources, 2000. Report to the General Assembly. New York: United Nations.

[3] ICRP (1991). Publication No. 60. Recommendations of the international commission on radiological protection (Vol. 21, pp. 1-3). Oxford: Annual Pergamon Press.

[4] Frančišković-Bilinski, S., Barišić, D., Vertaćnik, A., Bilinski, H., & Prohić, E. (2004). Characterization of tufa from the Dinaric Karst of Croatia: mineralogy, geochemistry and discussion of climate conditions. Facies, 50, 183–193. https://doi.org/10.1007/s10347-004-0015-8

[5] Pantelica, I. Georgescu, I., Murariu-Magureanu, M.D., Margaritescu, I., & Cincu, E. (2001). Thorium determination in intercomparison samples and in some Romanian building materials by gamma ray spectrometry. Radiation Protection Dosimetry, 97(2), 187–191.

[6] Turhan, Ş. (2010). Radioactivity levels of limestone and gypsum used as building raw materials in turkey and estimation of exposure doses. Radiation Protection Dosimetry, 140(4), 402–407.

[7] Kekelidze, N., Jakhutashvili, T., Tutberidze, B., Tulashvili, E., Akhalkatsishvili, M., Mtsariashvili, L., Chkhaidze, M., & Ambokadze, I. (2020). Radioactivity of rocks in several regions of East Georgia. In N. Todorović, & J. Nikolov (Eds.), Radionuclides: Properties, Behavior and Potential Health Effects (Chapter 3). Nova Science Publishers, Inc., Hauppauge, NY, USA.

[8] Kapanadze K., Magalashvili A., & Imnadze, P. (2019). Distribution of natural radionuclides in the soils and assessment of radiation hazards in the Khrami Late Variscan crystal massif (Georgia). Heliyon, 5, e01377. https://doi.org/10/1016/j.heliyon.2019.e01377.

[9] Anagnostakis, M.J., Hinis, E.P., Karangelos, D.J., Petropoulos, N.P., Rouni, P.K., Simopoulos, S.E., & Žinić, Z.S. (2001). Determination of depleted uranium in environmental samples by gamma-spectroscopic techniques. Archive of Oncology, 9(4), 231-236.

[10] Jubelt, R., & Schreiter, P. (1977). Rocks identification guide. Mir, Moscow.

[11] NuDat. National Nuclear Data Center. Brookhaven National Laboratory, USA. https://www.nndc.bnl.gov/nudat3/.

[12] Diab, H.M., Nouh, S.A., Hamdy, A., & El-Fiki, S.A. (2008). Evaluation of natural radioactivity in a cultivated area around a fertilizer factory. Journal of Nuclear and Radiation Physics, 3(1), 53-62.

[13] Caridi, F., Paladini, G., Gregorio, F., Lanza, S., Lando, G., Sfacteria, M., Tuccinardi, S., Venuti, M., Cardiano, P., Majolino, D., & Venuti, V. (2025). Natural radioactivity content and

radon exhalation rate assessment for building materials from the archaeological park of Tindari, Sicily, Southern Italy: A Case Study. Int. J. Environ. Res. Public Health, 22, 379. https://doi.org/10.3390/ijerph22030379.

[14] Amasi, A. I., Mtei, K. M., Nathan, I. J., Jodłowski, P., & Dinh, Ch. N. (2014) Natural radioactivity in Tanzania cements and their raw materials. Research Journal of Environmental and Earth Sciences, 6(10), 469 – 474.

[15] Gawad, A. E. A., Eliwa, H., Masoud, M. S., Khandaker, M. U., & Hanfi, M. Y. (2023). Assessment of the potential radiation hazards posed by Nubian sandstone, Egypt. Sci Rep, 13, 21202. https://doi.org/10.1038/s41598-023-47150-4.

[16] Agora, J. O., & Hashim, N. O. (2015). Assessment of radioactivity levels and risks due to different rock types from the KERIO Valley "High Background Radiation Area (HBRA)" of Kenya. Int. J. of Math. and Phys. Sciences Research, 3(1), 40 – 47.

[17] Oyeyemi, K. D., Aizebeokhai, A. P., Ekhaguere, O. A., Chinwuba, D. E., & Onumejor, C. A. (2019). Radiogenic components of limestone samples collected from Ewekoro SW Nigeria: Implications for public radiological health risks assessment and monitoring. Maced. J. Med. Sci., 7(19), 3287-3293. https://doi.org/10.3889/oamjms.2019.661.

[18] Najam, L. A., AL-Jomaily, F. M., & AL-Farha, E. M. (2011). Natural radioactivity levels of limestone rocks in northern Iraq using gamma spectroscopy and nuclear track detector. J. Radioanal. Nucl. Chem., 289, 709–715. https://doi.org/10.1007/s10967-011-1144-5