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Determination of Radioactivity in Soil Samples and Evaluation of Excess Lifetime Cancer Risk in Albania

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Abstract. The aim of this study was determination of natural (^{226}Ra, ^{232}Th, ^{40}K) and artificial (^{137}Cs) radioactivity levels in thirty soil samples collected from different locations in Albania. The obtained results provides essential information concerning reference baseline concentrations of natural and artificial radioactive isotopes and environmental pollution. The main contributors of natural radioactivity in soil are ^{226}Ra, ^{232}Th and ^{40}K. Artificial radionuclides can be also present such as ^{137}Cs, resulting from fallout from weapons testing or nuclear accidents. For determination of natural and artificial radionuclides concentration in the environment non-destructive measurement technique with High Pure Germanium detectors (HPGe) is used. The soil samples were collected at maximum depth of 10 cm. The average activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were found to be 22.18 ± 0.73 Bq kg^{-1}, 23.53 ± 0.93 Bq kg^{-1}, 326 ± 15 Bq kg^{-1} and 6.29 ± 0.45 Bq kg^{-1} in soil samples, respectively. In order to evaluate the radiological hazard of radioactivity, the total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR) have been calculated. The average absorbed dose rate D (nGy h^{-1}) in air at 1 m above ground level due to the ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the soil samples was 38.23 nGy h^{-1}. The calculated values of annual effective dose (AEDE) for the all soil samples ranged from 3.59 to 94.94 μSv y^{-1} with a mean of 46.89 μSv y^{-1}, which is lower than the world average value of 70 μSv y^{-1}. The average of excess lifetime cancer risk (ELCR) value was calculated to be 1.6×10^{-4}. Moreover compared to the World's average of 2.9×10^{-4}, the lifetime risk of cancer result are lower almost for all localities. Therefore, the all mean values of radiological hazard for all soil samples, were lower than the world average, it is safe for population living or other human activities with acceptable radiological risk.

Keywords: Radioactivity, gamma dose rate, radiological hazard, ELCR

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1. Introduction

Since formation of the Earth, radionuclides with long half-lives have been present in the Earth's crust. As a result of their radioactivity, determination of the activity levels of these radionuclides in environment is essential. Human beings are exposed to background radiation that comes both from natural and manmade sources. Radionuclides are everywhere, in the air, in the water, in the ground, in our houses, our bodies, etc.

The main contributors of natural radioactivity in soil are ^{226}Ra, ^{232}Th and ^{40}K, because the others terrestrial radionuclides are present in nature, but at such low levels that their contributions to the dose in the humans are small [1]. Artificial radionuclides can also be present such as ^{137}Cs, resulting from fallout from weapons testing or nuclear accidents as fission product. The radionuclides release from fission, by nuclear accident at town of Chernobyl, in Ukraine (April 1986) caused a significant increase of artificial radioactivity, especially of radionuclides of ^{137}Cs, ^{134}Cs, and ^{131}I. Nowadays, there are traces in the ecosystems only of ^{137}Cs due to its long half-life of 30.05 y [2].

Soil acts as a source of continuous radiation exposure to human and as a medium of migration for transfer of radionuclides to the biological system, hence causes the radiological contamination in the environment. On the other hand, storage reference data records on radionuclides is important for producing a radiation map of the country and ascertaining possible changes in environmental radioactivity caused by nuclear, industrial and other human activities [3]. These radionuclides are not uniformly distributed by nature of the process of the formation of the soils and can and change over time, thus knowledge of their distribution in soil in different places and time play an important role in radiation protection and measurement. Some soils are continuously fertilized, while others are mixed with minerals by physical and chemical process, so radionuclides are moving from one place to others [4].

The radionuclides, also are transferred from soil directly or indirectly to the plant and after in the food chain, that we are using every day. So, the radionuclide taken into the body through food chain or the air and the harm they can cause depends on the behavior of each one in the body [5]. In the case of radium that is taken into the body from foods or air, most of it leaves the body and only 20% of it remains in our body and accumulates in our bones. The radium has chemical structure similar of the calcium and both release from bones is very slow. So, they remain in our body for long time and therefore when they have high levels they become more harmful to the body. Even thorium that is taken into body through bloodstream accumulates on the bone surface and remain for a long time in our body. Of course, potassium or the others radionuclide can enter in the our body, but more important is level of the radioactivity and radiological evaluation through the effective doses [6, 7]. Measuring level of radioactivity can be used to evaluate, dose rates, the annual effective dose and the last excess lifetime cancer risks [8].

The aim of this study has been not only to determine activity concentration values of gamma emitting radionuclides, but also to evaluate radiological hazard parameters such as total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR). The radiological hazard parameters were calculated and compared to the data reported by UNSCEAR 2000 [9].

2. Materials and methods

2.1. Study area

The soil samples are collected from different locations of Albania, which is located on the Balkan Peninsula in South and Southeast Europe. Albania cover an area of 28,748 km^2, which has mostly lowlands in the western and central part, while hills and mountains in the north and northeast and a considerable coastline in the western part. The population of Albania is around 2.8 million and most of it lives in the western and central part. In Table 1, sample ID, city and geographical location of samples are presented. The study include thirty locations, in which the soil samples are collected by physics students in the area where they come from. In the future, we hope to have more soil samples in order to cover as large as possible area in Albania.

Sample ID	City	North	East
S1	Laç	41.6405556	19.7163889
S2	Lundër	41.2833333	19.8666667
S3	Sukth	41.3758333	19.5347222
S4	Peshkopi	41.6500000	20.3833333
S5	Maliq	40.7147222	20.6994444
S6	Pogradec	40.8877778	20.6750000
S7	Balldre i ri	41.8233333	19.6333333
S8	Sarandë	39.8722222	20.0169444
S9	Rrajcë,	41.0758333	20.5683333
S10	Sarandë	39.8722222	20.0011111
S11	Lushnjë	40.8736111	19.7808333
S12	Durrës	41.3330556	19.4494444
S13	Peqin	41.0461111	19.8058333
S14	Lushnjë	40.9357730	19.7145430
S15	Fier	40.7295875	19.5489844
S16	Lezhë	41.7794444	19.6394444
S17	Korçë	40.6383333	20.7677778
S18	Fushë Krujë	41.5025000	19.6972222
S19	Shkodër	42.0519667	19.4954333
S20	Kavajë	41.1691167	19.5601500
S21	Fishtë	41.8957167	19.6117500
S22	Zona e plazhit	41.3193000	19.4964000
S23	Kashar	41.3683333	19.6975667
S24	Rrogozhinë	41.0609833	19.6426333
S25	Plazhi Gjeneralit	41.1333333	19.4642778
S26	Bitinckë 1	40.6334631	20.9922334
S27	Bitinckë 2	40.6415832	20.9887783
S28	Bitinckë 3	40.6416113	20.9890002
S29	Bitinckë 4	40.6417784	20.9886111
S30	Bitinckë 5	40.6418897	20.9902786

Table 1**.** Geographical location of soil samples.

2.2. Sampling and sample preparation

The soil samples are taken in plastic bags, marking the place and coordinates for each of them and after were open on trays in the air for a period about one week in the laboratory conditions to remove a part of humidity. All samples were crashed, pulverized and sieved by a mesh to have average particle size lower than 1 mm, and then are homogenized. Samples were dried on oven at a temperature about 100 ^{0}C for 24 hours until constant mass was obtained and to remove as much as possible humidity. Each of them were transferred into a Marinelli beaker with

a volume of 500 ml, where net mass found by weighted accurately and finally hermetically sealed, in order to prevent leaks of radon gas ^{222}Rn. Soil samples were stored for 30 days before measurements in order to reach a secular equilibrium between ^{226}Ra and progeny.

2.3. Sample analysis

The radiation levels of soil samples were analyzed by gamma-ray spectrometry and were performed by using a detector p-type High Purity Germanium (HPGe) in the laboratory of Institute of Applied Nuclear Physics (IANP) in Tirana, Albania. Relative efficiency of detector is 40% and has Full Width at Half Maximum (FWHM) of 1.8 keV in 1332.5 keV from gamma emission of ^{60}Co. Soil samples spectra analysis were performed by using software Genie 2000 (Version 3.2.1). The Marinelli beaker after secular equilibrium were placed on the top of detector, and time of measurement for each sample was 86400 seconds. Energy calibration is performed using some point sources and absolute efficiency calibration in every peak is performed in the energetic range from 30 keV to 2000 keV, using Laboratory Sourceless Calibration Software (LabSOCS) [10]. The efficiency calibration curve were validated by using reference material supplied by the International Atomic Energy Agency (IAEA) and by the international participation in IAEA Worldwide Proficiency Test for environmental radionuclides [11].

The net area for every energy peak was calculated from total counts subtracting counts due to Compton scattering and other counts by background sources surrounding. The activity concentration A of every radionuclide, were calculated from the net area of the a certain peak by the using equation 1:

$$A = \frac{N_{net}}{\varepsilon(\mathrm{E}_\gamma) \cdot \mathrm{I}_\gamma \cdot \mathrm{t} \cdot \mathrm{m}} \qquad (1)$$

The activity concentration A and is expressed in Bq kg^{-1}, while N_{net} is the net peak area for radionuclide in the energy peak, $\varepsilon(\mathrm{E}_\gamma)$ is the measured counting efficiency of our detector in certain peak, I_γ is the intensity of the gamma line from the radionuclide expressed in percent, t is the counting live time and m is the dried sample mass in kilogram [12].

Minimum Detectable Activity (MDA) were calculated by using Currie equation 2 [13].

$$MDA = \frac{2.71 + 4.65\sqrt{N_B}}{\varepsilon(\mathrm{E}_\gamma) \cdot \mathrm{I}_\gamma \cdot \mathrm{t} \cdot \mathrm{m}} \qquad (2)$$

where, N_B is background counts under the corresponding peak, while the other terms entering the formula are mentioned above in the equation (1). The activity concentration and uncertainties of natural and artificial radionuclides of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the soil samples were determined. The activity concentrations were determined by activity concentration of daughter products in the case of ^{226}Ra and ^{232}Th and by main energy peak of radionuclides of ^{40}K and ^{137}Cs.

In this way, the activity concentration of ^{226}Ra was calculated by averaging activities of daughter radionuclides ^{214}Pb and ^{214}Bi. Activity concentration of ^{214}Pb was calculated using 241.99, 295.22 and 351.93 keV gamma lines and ^{214}Bi using 609.31 keV and 1120.29 keV. The activity of ^{232}Th was calculated using 338.4 and 911.2 keV gamma lines of ^{228}Ac. For ^{40}K and

^{137}Cs, the activity concentration was calculated using 1460.82 keV and 661.66 keV gamma lines. Gamma-ray intensities were taken from the Nuclide-LARA library [14].

3. Results and discussion

3.1. Activity concentrations in soil samples

The results of activity concentrations of natural radioactivity for radionuclides of ^{226}Ra, ^{232}Th, ^{40}K and artificial radionuclide of ^{137}Cs for thirty soil samples are presented in the Table 2. The activity concentrations for every radionuclide are expressed in Bq kg^{-1} and the uncertainty is within ±1σ. The results indicate that natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were present in all samples, whereas the presence of artificial radionuclide ^{137}Cs it is not found in all of them.

The range of activity concentration of ^{226}Ra is from 4.72 ± 0.22 Bq kg^{-1} to 63.0 ± 1.9 Bq kg^{-1}, for ^{232}Th from 0.65 ± 0.11 Bq kg^{-1} to 57.1 ± 1.9 Bq kg^{-1} and for ^{40}K is from 2.04 ± 0.96 Bq kg^{-1} to 662 ± 28 Bq kg^{-1}. The values of ^{137}Cs are below MDA of 0.20 Bq kg^{-1} for four soil samples presented in the end of the Table 2 until to 48.7 ± 3.0 Bq kg^{-1}, which is the higher value.

No.	**Activity Concentration (Bq kg^{-1} ± 1σ)**			
ID	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
S1	27.75 ± 0.92	27.1 ± 1.1	405 ± 18	2.99 ± 0.30
S2	24.95 ± 0.79	28.5 ± 1.0	428 ± 19	2.16 ± 0.18
S3	23.32 ± 0.79	29.0 ± 1.1	479 ± 21	1.53 ± 0.16
S4	41.1 ± 1.2	57.1 ± 1.9	565 ± 24	13.54 ± 0.86
S5	24.71 ± 0.81	31.6 ± 1.1	526 ± 23	9.72 ± 0.63
S6	24.13 ± 0.78	29.0 ± 1.1	465 ± 20	2.94 ± 0.22
S7	23.48 ± 0.76	24.53 ± 0.92	382 ± 17	1.49 ± 0.21
S8	63.0 ± 1.9	52.9 ± 1.7	332 ± 15	14.12 ± 0.89
S9	7.60 ± 0.37	7.17 ± 0.44	117.8 ± 5.9	48.7 ± 3.0
S10	38.4 ± 1.2	40.3 ± 1.4	280 ± 13	3.84 ± 0.29
S11	30.19 ± 0.94	38.3 ± 1.3	527 ± 23	0.75 ± 0.17
S12	21.53 ± 0.71	23.59 ± 0.89	425 ± 18	2.77 ± 0.21
S13	31.4 ± 1.0	36.8 ± 1.3	427 ± 19	4.79 ± 0.34
S14	16.92 ± 0.59	17.10 ± 0.72	300 ± 14	7.68 ± 0.50
S15	16.92 ± 0.60	17.52 ± 0.75	332 ± 15	6.27 ± 0.50
S16	19.27 ± 0.65	21.03 ± 0.82	332 ± 15	6.58 ± 0.44
S17	23.15 ± 0.75	22.10 ± 0.85	369 ± 16	21.1 ± 1.3
S18	27.42 ± 0.90	37.8 ± 1.4	662 ± 28	4.53 ± 0.33
S19	23.91 ± 0.76	25.9 ± 1.0	437 ± 18	6.84 ± 0.42
S20	11.63 ± 0.43	12.56 ± 0.53	184.3 ± 7.9	1.10 ± 0.08
S21	23.86 ± 0.76	28.0 ± 1.1	363 ± 15	6.37 ± 0.39
S22	18.53 ± 0.59	14.34 ± 0.62	352 ± 15	7.49 ± 0.46
S23	30.6 ± 1.1	32.1 ± 1.3	448 ± 19	3.88 ± 0.24
S24	17.26 ± 0.62	16.38 ± 0.67	283 ± 12	0.83 ± 0.06
S25	7.74 ± 0.27	8.53 ± 0.30	205 ± 14	5.98 ± 0.37
S26	4.72 ± 0.22	0.65 ± 0.11	8.0 ± 1.4	0.79 ± 0.1
S27	7.07 ± 0.27	0.73 ± 0.11	2.04 ± 0.96	< 0.20
S28	10.64 ± 0.38	8.7 ± 1.4	79.8 ± 4.1	< 0.20
S29	9.28 ± 0.34	5.4 ± 1.2	20.0 ± 1.7	< 0.20
S30	14.92 ± 0.47	11.31 ± 0.94	37.0 ± 2.2	< 0.20

Table 2. Activity concentration of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in thirty soil samples

As it seems from the Table 2, the higher values of the activity concentrations are those of natural radionuclides. As expected, gamma radiation from these natural radionuclides are the main

contributors source of radiation to the human. In addition, the activity concentration of ^{40}K is much higher compare with three other radionuclides of ^{226}Ra, ^{232}Th and ^{137}Cs.

The average values of activity concentrations calculated in our soil samples for ^{226}Ra, ^{232}Th ^{40}K and ^{137}Cs are 22.18 Bq kg^{-1}, 23.53 Bq kg^{-1}, 326 Bq kg^{-1} and 6.29 Bq kg^{-1}. These values of ^{226}Ra, ^{232}Th and ^{40}K are lower than the world average value of activity concentrations of 32 Bq kg^{-1}, 45 Bq kg^{-1} and 420 Bq kg^{-1} that are presented in Table 3 [9]. The average values of activity concentrations are comparable with values in selected regions neighboring countries. The activity concentration of ^{40}K can be higher, due to agricultural land by using of potassium fertilizers and also from accumulated soils because of geological origin. For ^{137}Cs values below MDA are in fact to minerals waste extracted from underground of the mine in Bitinckë, mixed with soils.

Country	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**	**References**
Italy	79	48	640	25	[15]
Turkey	85.75	51.08	771.57	236.38	[8]
Montenegro	41.1	45.8	499.8	95.2	[16]
North Macedonia	38.8	43.7	546	41.5	[17]
Serbia	45	50.3	650.5	10.2	[18]
Croatia	44.7	42.3	542	30.8	[19]
Greece	52	52	673		[20]
Kosovo*	22.3	21.1	358.2	12.9	[21]
UNSCEAR 2000	32	45	420		[9]
Albania	26.2	25.2	408.2		[22]
Albania	**22.18**	**23.53**	**326**	**6.29**	**Present study**

Table 3. Comparison of the average of activity of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in selected regions neighboring countries (Bq kg^{-1})

3.2. Evaluation of the radiological effects from soil samples

In order to evaluate the radiological hazard of radioactivity the total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR) have been calculated and are presented in Table 4. The absorbed dose rate in air on $1m$ above ground level by gamma radiation of radionuclides of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil samples was calculated. The absorbed dose rate D expressed in $nG\ h^{-1}$, was calculated from equation 3 [8, 9],

$$D(nG\ h^{-1}) = C_{Ra}A_{Ra} + C_{Th}A_{Th} + C_K A_K + C_{Cs}A_{Cs} \qquad (3)$$

where, A_{Ra}, A_{Th}, A_K, and A_{Cs} are the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, while C_{Ra}, C_{Th}, C_K and C_{Cs} are conversion factors of the dose rate per unit of activity concentration of radionuclides, respectively. Conversion factors for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were taken by UNSCEAR 2000. The range of dose rate is from 2.93 nGy h^{-1} to 77.41 nGy h^{-1} and the average value is 38.23 nGy h^{-1}, which is lower than the world average value of 60 nGy h^{-1} [9].

The annual effective dose equivalent (AEDE) by gamma radiation of radionuclides of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil samples were found. The value of AEDE is expressed in μSv y^{-1} and was calculated from equation 4 [9],

$$AEDE(\mu Sv\ y^{-1}) = D(nG\ h^{-1}) \cdot 0.7(Sv\ Gy^{-1}) \cdot 8760(h\ y^{-1}) \cdot 0.2 \qquad (4)$$

where, D is absorbed dose rate, while 0.7 is the conversion coefficient from absorbed dose in air to effective dose received for adults, by UNSCEAR reports [9, 23]. The annual effective dose equivalent was evaluated for a year, where number of hours is 8760 and 0.2 is the outdoor occupancy factor. The range of the AEDE is from 3.59 μSv y^{-1} to 94.94 μSv y^{-1} and the average value is 46.89 μSv y^{-1}, which is lower than the world average value of 70 μSv y^{-1} [9].

ID	$D(nGy\ h^{-1})$	$AEDE(\mu Sv\ y^{-1})$	$ELCR(x10^{-4})$
S1	46.17	56.62	2
S2	46.68	57.25	2
S3	48.30	59.23	2.1
S4	77.41	94.94	3.3
S5	52.73	64.67	2.3
S6	48.15	59.05	2.1
S7	41.66	51.09	1.8
S8	75.34	92.39	3.2
S9	14.21	17.43	0.6
S10	53.89	66.09	2.3
S11	59.06	72.43	2.5
S12	41.99	51.50	1.8
S13	54.65	67.02	2.3
S14	30.88	37.87	1.3
S15	32.44	39.78	1.4
S16	35.67	43.74	1.5
S17	40.05	49.12	1,7
S18	63.23	77.55	2.7
S19	45.07	55.27	1.9
S20	20.67	25.36	0.9
S21	43.27	53.07	1.9
S22	32.14	39.41	1.4
S23	52.29	64.13	2.2
S24	29.71	36.432	1.3
S25	17.45	21.40	0.8
S26	2.93	3.59	0.1
S27	3.29	4.65	0.2
S28	13.50	16.56	0.6
S29	6.36	10.26	0.4
S30	15.27	18.72	0.7

Table 4. Absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) by gamma radiation of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil samples was found. The value of ELCR was calculated from equation 5 [24],

$$ELCR = AEDE(\mu Sv\ y^{-1}) \cdot AL \cdot RF \qquad (5)$$

where, AEDE is the annual effective dose equivalent, while AL is the average lifetime about 70 years. RF is risk factor expressed in Sv^{-1} and for stochastic effects has value 0.05 for the public [25]. The range of the ELCR varies from $1 \cdot 10^{-5}$ to $3.3 \cdot 10^{-4}$ and the average value is $1.6 \cdot 10^{-4}$. The

average value of ELCR due to the presence natural and artificial radionuclides in the samples was $1.6 \cdot 10^{-4}$, which is lower than the world average value of $2.9 \cdot 10^{-4}$ [9]. The radiological hazard of radioactivity, the total absorbed dose rate, the annual effective dose equivalent and excess lifetime cancer risk have been evaluated for thirty soil samples. The all average values of radiological hazard for all soil samples, were lower than the world average value. It is safe for population living or other human activities with acceptable radiological risk.

4. Conclusion

In this paper, the natural and artificial radioactivity of 30 soil samples in Albania is presented. Also, radiological hazard of radioactivity, the total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR) have been evaluated. The radiological hazard parameters from soil samples were evaluated and compared to the data reported by UNSCEAR 2000.

The average values of activity concentration calculated in the soil samples for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs are 22.18 Bq kg^{-1}, 23.53 Bq kg^{-1}, 326 Bq kg^{-1} and 6.29 Bq kg^{-1}. These values are lower than the world average values of 32 Bq kg^{-1}, 45 Bq kg^{-1} and 420 Bq kg^{-1}.

The average values of the absorbed dose rate, the annual effective dose equivalent and the excess lifetime cancer risk were found to be: 38.23 nGy h^{-1}, 46.89 μSv y^{-1} and $1.6 \cdot 10^{-4}$.

Radiological hazard of the activity concentrations, the total absorbed dose rate, the annual effective dose equivalent and excess lifetime cancer risk have been evaluated and the all average values of them, were lower than the world average value. Therefore, it is safe for population living or other human activities with acceptable radiological risk.

The obtained data provide us the baseline levels of natural and artificial radioactivity, evaluation of the radiological effect from soil samples, also a background information for future research on the others soil samples, for radiological protection of the human and safety of our health.

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