ECOLOGIA BALKANICA
2024, Vol. 16, Issue 1 June 2024 pp. 102-111

Investigation of the radioactivity of soils collected from Shumen Plateau Nature Park, Bulgaria

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Abstract. The aim of the present study was to evaluate the levels of radioactivity, radiological doses and the increased lifetime risk of cancer of radionuclides contained in soils collected from the territory of Shumen Plateau Nature Park, (North-Eastern Bulgaria). A gamma spectrometric system was used to estimate the natural radionuclides. The average values for the specific activities for ^{238}U, ^{232}Th, ^{40}K measured in soils were 28.65 ± 4.30 Bq/kg, 18.86 ± 2.83 Bq/kg and 609.09 ± 91.36 Bq/kg, respectively. Only the average value for ^{40}K was higher than the documented worldwide values of, respectively 35 Bq/kg, 30 Bq/kg and 400 Bq/kg. We calculated the average values for assessing radiological hazards: radium equivalent activity 102.52 Bq/kg, external hazard index 0.28 Bq/kg, internal hazard index 0.35 Bq/kg, absorbed gamma dose rate in air 52.04 nGy/h, annual effective dose rate 0.32 mSv/y, total excess lifetime cancer risk 1.95×10^{-3} and radioactivity level index 0.79 mSv. The average values we obtained for radiological indices and specific activities are lower than the global average values quoted in UNSCEAR 2000.

Key words: radioactivity, soil, gamma ray spectrometry, radiation hazard indices, Shumen plateau.

Introduction

Both radionuclides of natural origin and artificial radionuclides resulting from human activity can be registered in the environment. Various factors may affect the movement of radionuclides in different parts of the earth. These factors can be divided into biotic and abiotic factors. The movement of radionuclides is mainly due to physical and mechanical processes in the atmosphere, lithosphere and hydrosphere. The most important repository for radionuclides in the natural environment is the soil (Seaman & Roberts, 2012). The role of the soil in relation to the presence and migration of radionuclides is related to the adsorption of a large part of the artificial radionuclides, reducing their activity for the root systems of plants (Iurian et al., 2015). On the other hand, the radionuclides are often fixed in the solid phase of the soil, which leads to long-term retention of radionuclides in the upper soil layer. The content of natural radionuclides in the soil depends on the type of the underlying rocks, as well as on the ongoing physicochemical processes (Vassilev, 2005).

About 80% of the total radiation that a human may receive in one year is due to radionuclides of terrestrial origin, found in soils and rock masses (IAEA, 1996). Among natural radionuclides in the environment, ^{40}K is the most abundant, as well as radionuclides from the two natural radioactive families with progenitors ^{238}U and ^{232}Th, which are present in the earth's crust (UNSCAER, 1993). Therefore, materials such as soil and building materials that are of natural origin may be the main sources of external radiation exposure of people in the environment. Studies involving dose rate estimation are essential, as they provide information on lifetime cancer risks (Ghias et al., 2021; Taskin et al., 2009). In the recent years, indices related to the amount of radionuclides entering the human body are increasingly used to assess the risk of internal and external exposure of a person (Raghu et al., 2015). According to the reports of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the radioactivity due to natural sources in the environment is equal to 2.4 mSv/y, and the share of anthropogenic sources was estimated at 0.8 mSv/y (Dhawal et al., 2013; UNSCEAR, 2000).

In the modern society, regular visits to nature parks are considered a good opportunity to achieve a healthy lifestyle. Koynova et al. (2019) analyzed the qualities of the Shumen Plateau Nature Park as a recreation site. Despite the intensive research on the conditions in the Nature Park which was conducted to date (Koleva et al., 2018, 2023; Koynova, 2018; Koynova & Koleva, 2021), data concerning the radiation background level are rather scarce. The purpose of the present study is to assess the radiation risk and the safety of the population considering that the territory of Shumen Plateau Nature Park is intensively used for sports and tourism (Koynova et al., 2019).

Materials and Methods

Shumen Plateau Nature Park is located in the southeastern part of the Mysia Plain. It includes the largest and highest southeastern part of the Shumen Heights. The area of Shumen Plateau Nature Park is 73 km^2. The highest point of the park, as well as of the Shumen Plateau, is peak Tarnov tabia (501.9 m). It is located in the western part of the Park. The karst, which defines the features of the natural environment, is very typical for the area. The region is characterized by 4 soil types and 13 subtypes and genera. The soils are humus-carbonate, dark gray, gray and light gray forest in the following percentage ratio 52%, 37%, 5.7% and 1.1%. The remaining 3.8% are rocks, unforested areas and others (Peichev & Radoslavova, 1998).

Sampling and radiometric analysis

19 soil samples were collected from various places in Shumen Plateau Nature Park, located along its entire length (the locations are presented in Fig. 1). The samples were taken from the upper soil layer to a depth of 30 cm. The mass of each analyzed sample is approximately 60 g.

Fig. 1. Sampling locations of Shumen Plateau Nature Park.

The analysis of the samples was carried out by a system for low-background gamma spectrometric measurements, which included: a semiconductor detector with a crystal volume of 60 cm^3, an operating voltage of 1 kV and a relative efficiency of 4.5% for the gamma line of ^{137}Cs with energy of 661.66 keV; a preamplifier, an amplifier, analog to digital converter with integrated multichannel analyzer (Analog to Digital Converter with integrated Multichannel Analyzer ADC-MCA) and a computer as a visualization device. The visualization of the spectra was performed with the software product SpectLab, and the processing of the gamma spectra with the program Anges.

Radiological parameters

Radium equivalent activity (Ra_{eq}).

This factor was applied to assess the radiological hazard of environmental samples. It was calculated by formula (Beretka & Mathew, 1985; UNSCEAR, 1988; UNSCEAR, 2000):

$$\boldsymbol{Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K}$$

where A_U, A_{Th}, and A_K were the specific activities of ^{238}U, ^{232}Th and ^{40}K.

External hazard index H_{ex} and internal hazard index H_{in}.

The indices are related to the danger of external (H_{ex}) or internal (H_{in}) irradiation of organisms. It is assumed that the same decay rate would be obtained from 370 Bq/kg for external irradiation and 185 Bq/kg for internal irradiation for ^{226}Ra, 259 Bq/kg for ^{232}Th and 4810 Bq/kg for ^{40}K present in the sample (Beretka & Mathew, 1985):

$$\boldsymbol{H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}}$$

$$\boldsymbol{H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}}$$

Absorbed dose rate

Due to the uniform distribution of the natural radio-nuclides ^{238}U, ^{232}Th and ^{40}K at 1 m above the Earth's surface, the outdoor absorbed dose rate in air can be estimated by the formula (UNSCEAR, 1988):

$$\boldsymbol{D_{out} = 0.436A_U + 0.599A_{Th} + 0.0417A_K}$$

Yearly effective dose equivalent

Assessment of the annual individual effective dose received outdoors can be made by taking into account the conversion factor from of the absorbed dose rate in air to the effective dose and the outdoor activity factor.

According to the reports of UNSCEAR (1993, 2000), the following conversion factors were used: 0.7 Sv/Gy was used to cover the absorbed dose in air to the effective dose received by an individual annually. It is considered that people spend outdoors about 20% of their time, therefore for the outdoor exposure occupancy factor was taken as 0.2. The annual individual effective dose E_{out} (mSv/y) outdoors can be calculated by the following formula:

$$\boldsymbol{E_{out}\left[\frac{mSv}{y}\right] = D_{out}\left[\frac{nGy}{h}\right] + 8760[h] * 0.2 * 0.7\left[\frac{Sv}{Gy}\right] = D_{out}1.226\,[\mu Sv]}$$

Absorbed gamma dose rate in air (ADRA)

The total absorbed dose of gamma radiation due to natural radionuclides outdoors at 1 m above ground level gives us information about the effects of gamma radiation from ambient radioactive sources on human health (Cengiz et al., 2018).

For the calculation of ADRA are used the specific activities ^{40}K, ^{232}Th and ^{238}U, as well as conversion factors (UNSCEAR, 2000):

$$\boldsymbol{ADRA = 0.461A_U + 0.623A_{Th} + 0.0417A_K}$$

Annual effective dose rate (AEDR)

Absorbed gamma dose в nGy/h may be converted in AEDR in mSv/y (UNSCEAR, 2000):

$$\boldsymbol{AEDR\left[\frac{mSv}{y}\right] = T.Q.ADRA.10^{-6}}$$

Radioactivity level index (I_γ)

For calculation of the values of Radioactivity level index (I_γ) are used the specific activities of ^{40}K, ^{232}Th and ^{238}U divided by the coefficients for ^{40}K, ^{232}Th and ^{238}U (NEA-OECD, 1979):

$$\boldsymbol{I_\gamma = \frac{A_u}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}}$$

Results and Discussion

The results for the specific activities in Bq/kg for ^{40}K, ^{232}Th and ^{238}U, obtained after the gamma spectrometric and statistical analysis, are presented in Table 1.

Table 1. Data from gamma spectrometric and statistical analyzes of radionuclide concentrations in soil from Shumen Plateau Nature park for ^{40}K, ^{232}Th and ^{238}U.

Variables	^{238}U	^{232}Th	^{40}K
Minimum	15.72	11.13	336.05
Maximum	35.39	22.37	877.02
Median	28.11	20.04	602.10
Mean	28.65	18.86	609.09
Std. Deviation	5.37	3.21	128.55
Mode	26	21	600
Skewness	- 0.93	- 1.36	0.02
Kurtosis	0.79	1.03	0.18
Pearson correlation	0.30	- 1.09	0.16

Geostatistical analysis was performed on the data obtained from the gamma spectrometric analysis, applying the Kriging technique. Via Surfer (Version 20.1.195) software, maps of the distribution of the specific activities of the radionuclides ^{40}K, ^{232}Th and ^{238}U have been performed, relative to the GPS coordinates of the sampling points (Fig. 2).

The statistical theory allows for the available spatial relationships to be used to interpolate values at locations, where sampling was not possible. The distribution maps clearly show the places with the highest and lowest concentration for the respective radionuclide.

The specific activity for ^{238}U was minimal in sample S14 15.72 ± 2.36 Bq/kg and maximal for sample S18 35.39 ± 5.31 Bq/kg, close to the World's average 35 Bq/kg (UNSCEAR, 2000) and average for Bulgaria 40 Bq/kg (UNSCEAR, 2000).

The specific activity calculated for ^{232}Th was minimal in sample S3 (11.13 ± 1.67 Bq/kg) and maximal in sample S7 (22.37 ± 3.36 Bq/kg), thus lower when compared to the average value of 30 Bq/kg reported by the World's and Bulgaria (UNSCEAR, 2000).

The highest concentration for ^{40}K was measured in sample S6 877.02 ± 97.96 Bq/kg, and the lowest concentration - in sample S3 336.05 ± 82.58 Bq/kg.

According to UNSCEAR (2000), the average value for ^{40}K for Bulgaria and the world is 400 Bq/kg, with values for Bulgaria ranging from 40 to 800 Bq/kg (UNSCEAR, 2000).

Statistical analysis was performed using the IBM SPSS Statistics (Version 19) program. Frequency distributions of the specific activities were obtained, which are graphically presented in Fig. 3. The percentage distribution of the specific activities obtained for ^{40}K, ^{232}Th and ^{238}U is well interpreted from the obtained graphs:

- ✓ In the interval from 25 to 35 Bq/kg fall 84.2% of the specific activities determined for ^{238}U;
- ✓ In the interval from 17.5 to 22.5 Bq/kg fall 79% of the specific activities determined for ^{232}Th;
- ✓ In the interval from 400 to 800 Bq/kg fall 89.5% of the specific activities defined for ^{40}K.

The data on the specific activities of ^{40}K, ^{232}Th and ^{238}U are presented by U-Th-K ternary diagram (Fig. 4). The ratio of the specific activities in all samples is preserved and all points fall in the same place – in the right top of the triangle.

The calculated coefficients related to the radiation risk and the average, minimum and maximum values for 19 soil samples are presented in Table 2.

The obtained average values for radiological parameters are compared with the recommended world standards. The calculated average value for *ADRA* is 52.04 nGy/h, for *AEDR* is 0.32 mSv/y, for Ra_{eq} is 102.52 Bq/kg, for H_{ex} is 0.28 Bq/kg, for H_{in} is 0.35 Bq/kg, for $ELCR_{tot}$ is 1.95 x 10^{-3} and for I_γ is 0.79 mSv. Those values are below the world's average ones - 59 nGy/h, 0.48 mSv/y, 370 Bq/kg, 1 Bq/kg, 1 Bq/kg, 0.05, 1 mSv, respectively (UNSCEAR, 2000).

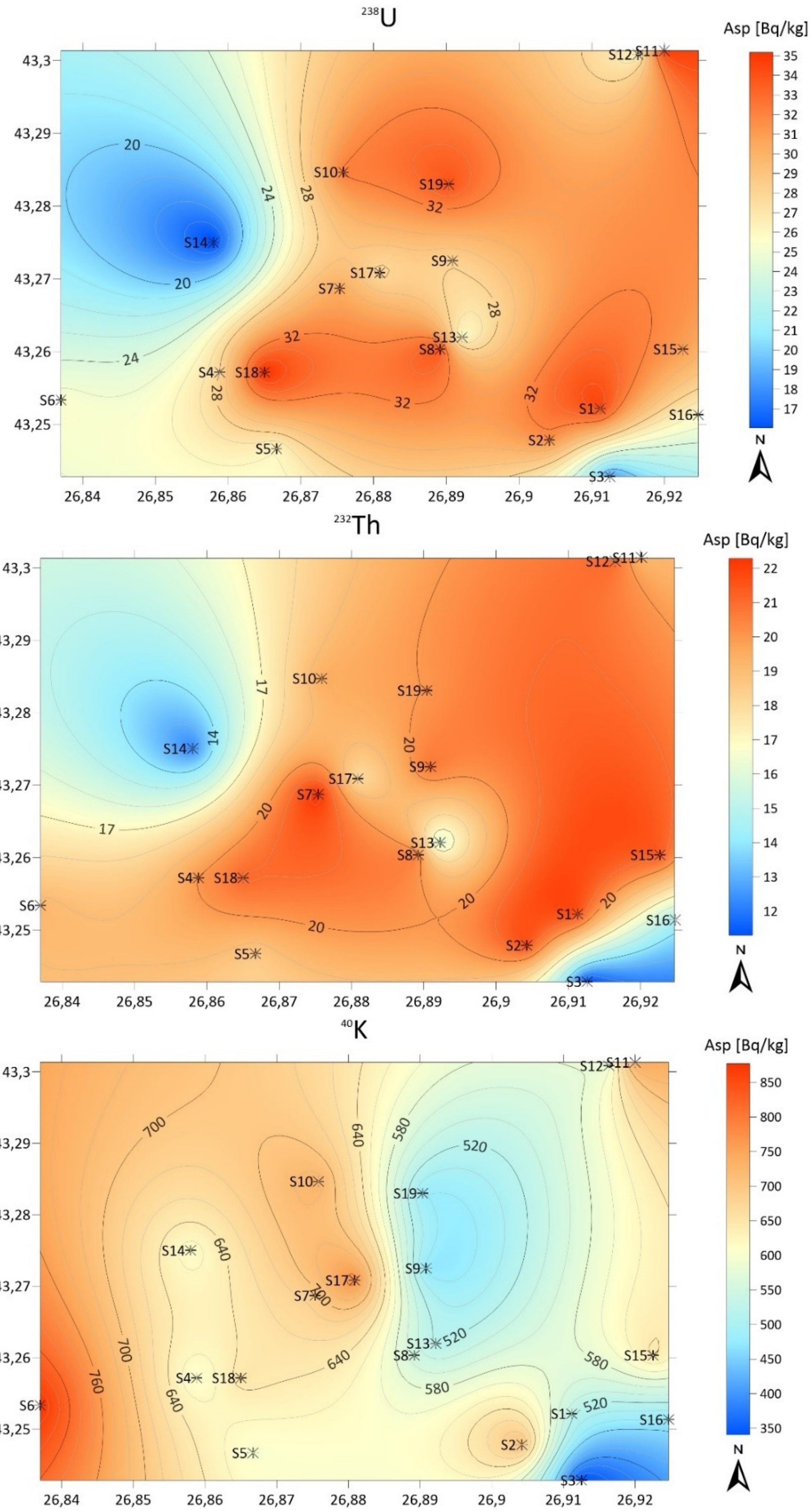

Fig. 2. Maps of distribution of ^{40}K, ^{232}Th and ^{238}U and GPS coordinates of the sample points.

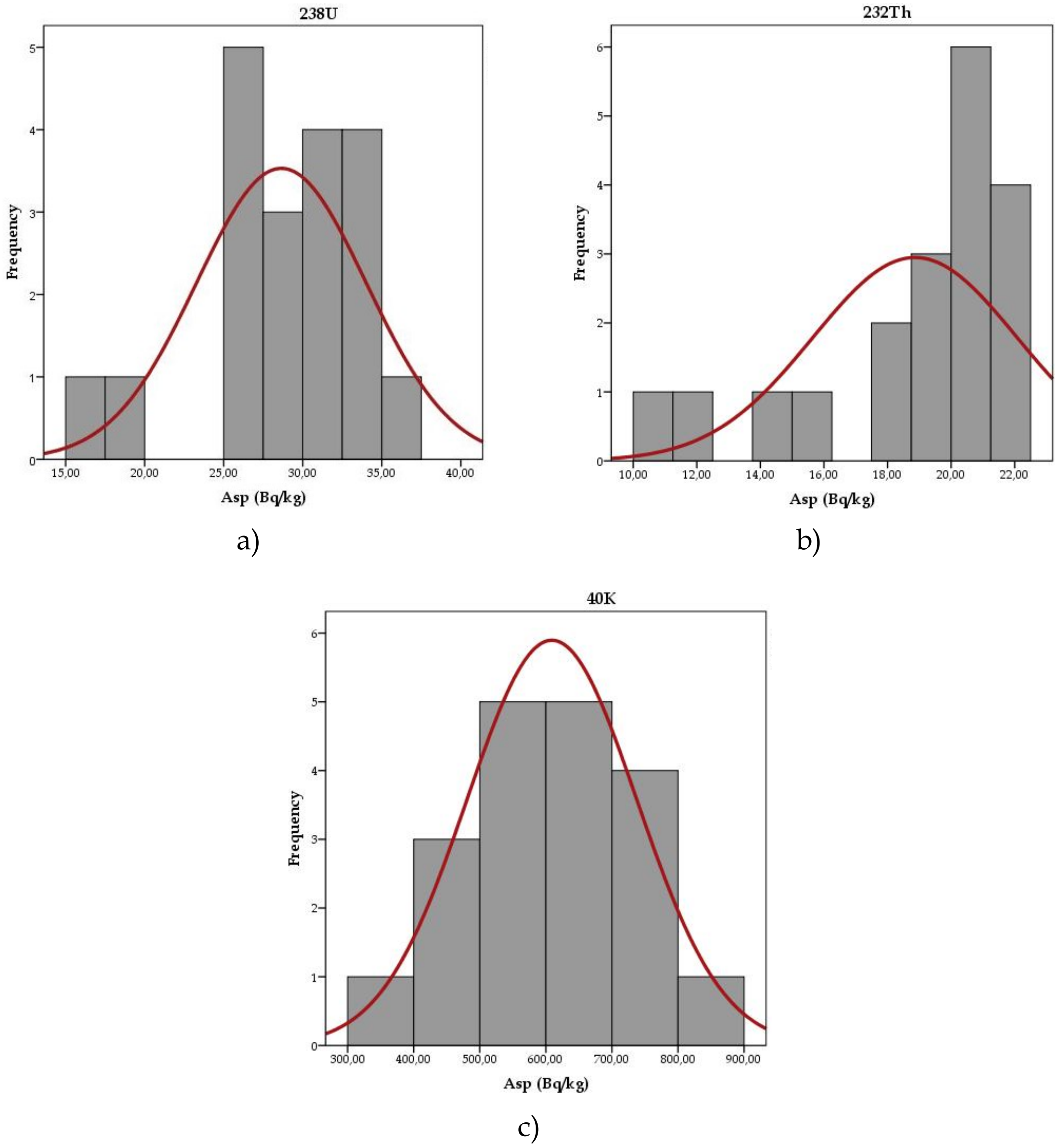

Fig. 3. Frequency distribution of: a) ^{238}U, b) ^{232}Th, c) ^{40}K

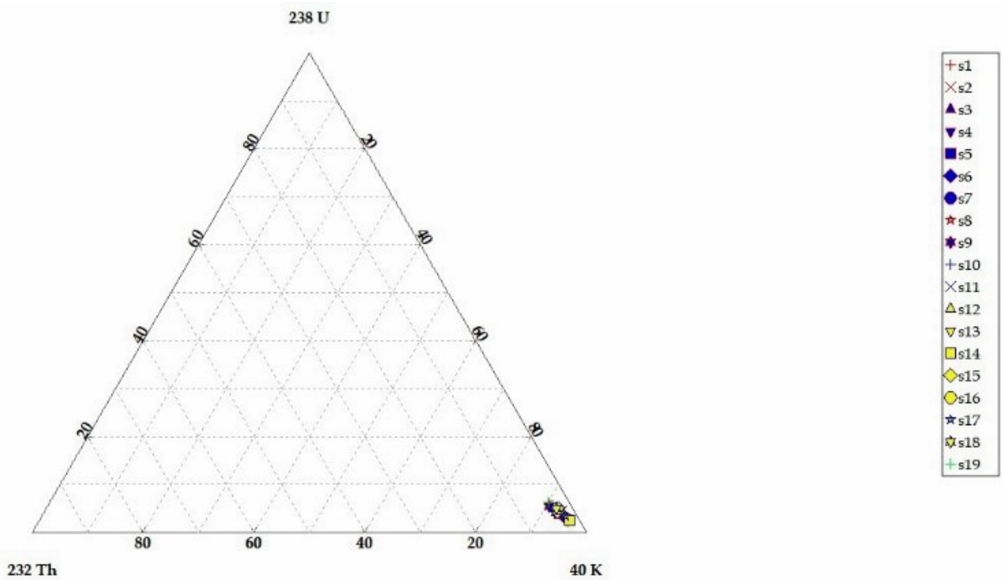

Fig. 4. Frequency distribution of: a) ^{238}U, b) ^{232}Th, c) ^{40}K.

Table 2. Absorbed gamma dose rate in air, annual effective dose rate, radium equivalent activity, external hazard index, internal hazard index, absorbed dose rate, yearly effective dose, equivalent total excess lifetime cancer risk, radioactivity level index in soil samples from Shumen Plateau Nature Park. World's average values are also reported.

Sampling Code	ADRA [nGy/h]	AEDR [mSv/y]	Ra_{eq} [Bq/kg]	H_{ex} [Bq/kg]	H_{in} [Bq/kg]	D_{out} [nGy/y]	E_{out} [mSv/y]	I_{γ} [mSv]	$ELCR_{tot}$ x 10^{-3}
S1	52.96	0.32	105.87	0.29	0.38	49.89	0.06	0.80	1.98
S2	59.50	0.36	116.84	0.32	0.40	56.16	0.07	0.90	2.22
S3	30.20	0.19	59.97	0.16	0.21	28.60	0.04	0.46	1.13
S4	52.45	0.32	102.77	0.28	0.35	49.19	0.06	0.79	1.94
S5	49.72	0.30	98.93	0.27	0.34	47.57	0.06	0.76	1.88
S6	62.48	0.38	120.39	0.33	0.39	59.15	0.07	0.95	2.33
S7	58.72	0.36	115.32	0.31	0.39	55.32	0.07	0.88	2.19
S8	52.93	0.32	105.45	0.28	0.38	49.95	0.06	0.80	1.98
S9	46.79	0.29	94.16	0.25	0.33	44.38	0.05	0.71	1.76
S10	58.39	0.36	116.20	0.31	0.40	56.12	0.07	0.90	2.22
S11	58.97	0.36	119.44	0.32	0.42	57.64	0.07	0.92	2.29
S12	50.93	0.31	101.66	0.27	0.35	48.60	0.06	0.78	1.92
S13	44.53	0.27	88.19	0.24	0.31	42.27	0.05	0.67	1.67
S14	43.52	0.27	80.09	0.22	0.26	39.57	0.05	0.63	1.56
S15	56.09	0.34	111.05	0.30	0.38	53.13	0.06	0.85	2.10
S16	44.32	0.27	83.40	0.23	0.30	39.89	0.05	0.64	1.58
S17	56.24	0.34	112.94	0.31	0.38	55.12	0.07	0.88	2.18
S18	59.02	0.36	115.61	0.31	0.41	55.17	0.07	0.88	2.19
S19	51.04	0.31	99.65	0.27	0.36	46.87	0.06	0.75	1.86
mean	52.04	0.32	102.52	0.28	0.35	49.19	0.06	0.79	1.95
min	30.20	0.19	59.97	0.16	0.21	28.60	0.04	0.46	1.13
max	62.48	0.38	120.39	0.33	0.42	59.15	0.07	0.95	2.33
World's average (UNSCEAR, 2000)	59	0.48	370	1	1	-	-	1	50

In Table 3, the Pearson's coefficients are presented, which indicate the relationship between the individual quantities. The coefficient was calculated for 12 radioactive variables. All the obtained values are positive numbers, which shows that the quantities have a linear relationship with each other. One value also falls in the interval from 0.3 to 0.5 (moderate correlation) and it is for ^{40}K and ^{232}Th. Four values fall in the interval from 0.5 to 0.7 (significant correlation), from 0.7 to 0.9 (high correlation) 24 coefficients fall and the most coefficients (48) fall into the group of very high correlation (from 0.9 to 1). A total of 93% of the obtained values fall into the interval for high and very high correlation between parameters.

A cluster analysis was performed on 12 parameters, the data from which are presented in Fig. 5. Cluster I consists of two sub clusters, including AEDR, H_{ex}, H_{in}, E_{out}, I_{γ}, ELCR, ADRA and D_{out} in one, and Ra_{eq}, Th and U in the other. This implies that a large quantity of natural radioactivity is due to gamma-emitting radionuclides from ^{232}Th and ^{238}U families. Cluster II includes ^{40}K, which suggests that the concentration of the isotope does not have a significant impact on the formation of the natural radioactivity.

Table 3. Coefficients obtained by Pearson's correlation.

	^{238}U	^{232}Th	^{40}K	ADRA	AEDR	Ra_{eq}	H_{ex}	H_{in}	D_{out}	E_{out}	I_Y	ELCR
^{238}U	*1*											
^{232}Th	*0.811*	1										
^{40}K	*0.253*	*0.394*	*1*									
ADRA	*0.686*	*0.777*	*0.852*	*1*								
AEDR	*0.686*	*0.777*	*0.852*	*1*	*1*							
Ra_{eq}	*0.733*	*0.811*	*0.825*	*0.991*	*0.991*	1						
H_{ex}	*0.733*	*0.812*	*0.825*	*0.991*	*0.991*	*1*	1					
H_{in}	*0.844*	*0.856*	*0.718*	*0.964*	*0.964*	*0.983*	*0.983*	1				
D_{out}	*0.731*	*0.810*	*0.827*	*0.989*	*0.992*	*1*	*1*	0.978	1			
E_{out}	*0.731*	*0.810*	*0.827*	*0.989*	*0.992*	*1*	*1*	0.978	1	1		
I_Y	*0.676*	*0.769*	*0.870*	*0.993*	*0.993*	*0.996*	*0.996*	*0.965*	0.997	0.997	1	
ELCR	*0.689*	*0.770*	*0.864*	*0.993*	*0.993*	*0.997*	*0.997*	*0.970*	*0.998*	*0.998*	*1.000*	1

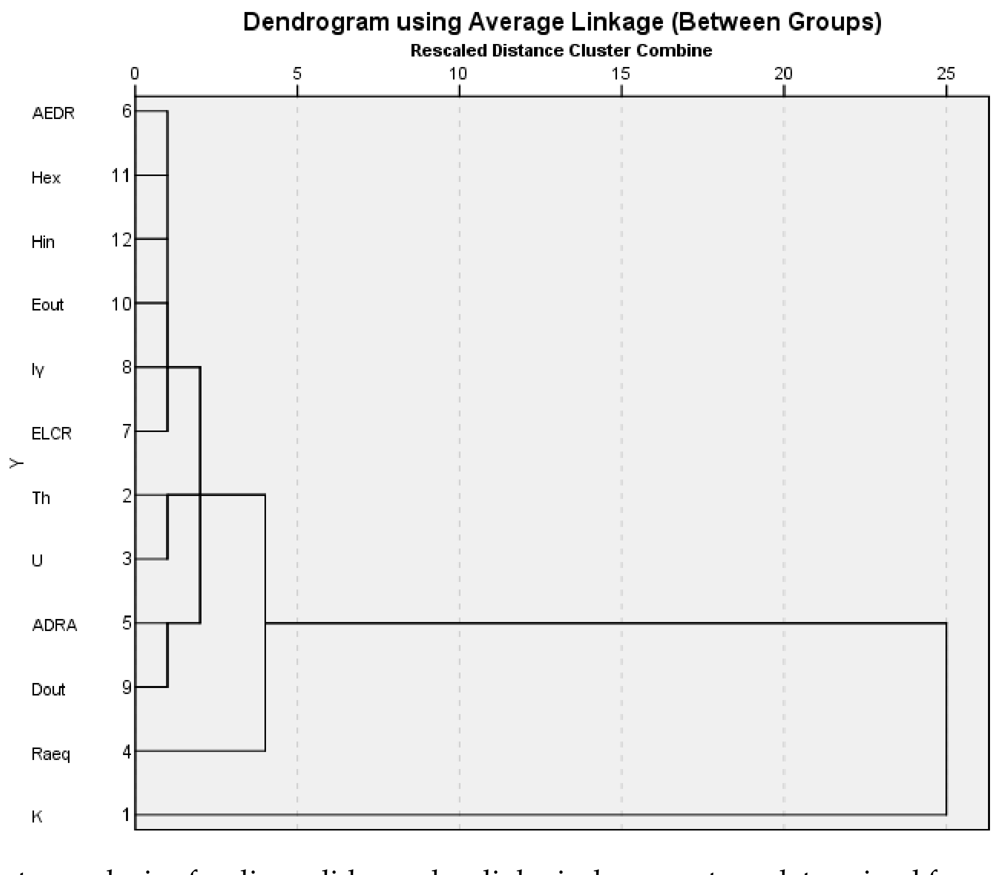

Fig. 5. Cluster analysis of radionuclides and radiological parameters, determined for soil samples collected from Shumen Plateau Nature Park.

Conclusions

The present work represents results related to the assessment of the radiation risk of soils collected from the Shumen Plateau National Park. All calculated radiation coefficients are lower than the internationally quoted values according to UNSCEAR (2000).

The results obtained after the cluster analysis and Pearson's correlation analysis show that radionuclides from the two natural radioactive families with progenitors ^{238}U and ^{232}Th mainly contribute to radioactivity on the territory of Shumen Plateau Nature Park.

The results of the present study can be used for a more accurate global picture of the radioactivity in northeastern Bulgaria.

Acknowledgments: The measurements were made with the financial support of project RD-08-113/20.02.2023, of the "Episkop Konstantin Preslavski" University of Applied Sciences. This research is supported by the Bulgarian Ministry of Education and Science under the National Program "Young Scientists and Postdoctoral Students – 2".

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Received: 03.11.2023
Accepted: 20.05.2024