RAP CONFERENCE PROCEEDINGS, VOL. 10, PP. 131–138, 2025
ISSN 2737-9973 (ONLINE) | DOI: 10.37392/RAPPROC.2025.23
RAP-PROCEEDINGS.ORG

U-238 AND Th-232 IN SOIL AND SAND IN MONTENEGRO: CORRELATIONS AND DOSE RATES

Nevenka M. Antović*

University of Montenegro, Faculty of Natural Sciences and Mathematics, Podgorica, Montenegro

Abstract. *Activity concentrations of ^{238}U and ^{232}Th in soils and beach sands of Montenegro were analyzed to evaluate their correlations, as well as absorbed dose rates in air coming from these two natural radionuclides. The analysis was based on the data obtained in the previous period using in situ gamma spectrometry and standard laboratory HPGe spectrometry. This paper is focused on 20 sand points at the beaches along the Coast of Montenegro, 42 soil points at the territory of entire Montenegro, and additional 12 soil points at the territory of Nikšić – the second largest town in the country, having a complex geological base. Uranium-238 in soil was found to range from 7 to 166 Bq/kg with an average of 29.3 Bq/kg (Montenegro) and from 39.7 to 112 Bq/kg with an average 59.5 Bq/kg (Nikšić), as ^{232}Th ranged from 9.3 to 74 Bq/kg with an average of 23.7 Bq/kg (Montenegro) and from 33.9 to 78.6 Bq/kg with an average of 54.4 Bq/kg (Nikšić). Normality of the radionuclide activity distributions is tested by the Q-Q plots and Kolmogorov-Smirnov and Shapiro-Wilk tests, whilst the linear regression analysis showed that 64.8% of the variation in ^{232}Th activity concentration in soils of Montenegro could be explained by ^{238}U as a predictor. The ^{232}Th and ^{238}U activity concentrations in soil at the entire Montenegro territory are found to be positively correlated (Pearson correlation coefficient: r = 0.805), but not in the Nikšić soil, where statistically significant correlation is found for ^{232}Th and ^{226}Ra (r = 0.856). Very strong positive correlation between ^{232}Th and ^{40}K was observed for beach sands, where the ^{238}U (and ^{226}Ra), ^{232}Th and ^{40}K activity ranged up to 16 Bq/kg, 16.6 Bq/kg and 305 Bq/kg, respectively. Absorbed dose rate in air coming from ^{238}U in soil has average values 13.5 and 27.5 nGy/h for Montenegro and Nikšić, respectively, and that coming from ^{232}Th – 14.3 and 32.9 nGy/h, respectively. The dose rates from ^{238}U and ^{232}Th in beach sands are lower, not exceeding 7.4 nGy/h (^{238}U) and 10 nGy/h (^{232}Th).*

Keywords: *soil and sand, natural radionuclides, activity concentration, linear correlation, absorbed dose rate*

1. INTRODUCTION

Natural radionuclides of terrestrial origin, such as ^{40}K and radionuclides from the ^{238}U and ^{232}Th series, are present in various amounts in all media in the environment [1]. Human exposure to external sources of ionizing radiation is mainly caused by gamma radiation from decays of these radionuclides. Therefore, investigation of their concentrations in different compartments of the environment is important from the radioecological and radiation protection viewpoint.

Activity concentrations of natural radionuclides in soils have been extensively studied worldwide to assess external exposure outdoors [1], [2]. Natural radionuclides in soil also contribute to the internal exposure of the human population through ingested food, since they may get transferred to plants during mineral uptake, accumulate in various parts, including the edible ones [3].

It is important to point out that the level of natural radionuclides in soil is related to the rock from which the soil originates, but also to the soil genesis. Hence, the knowledge of activity concentrations of radionuclides in soil and rock samples can be a powerful tool for discriminating different lithologies [4]. The relationships between the radionuclides and soil properties can be used to assess the pedogenic effect on the pattern distribution of natural radionuclides [5], etc.

Systematic measurements of naturally occurring radionuclides in Montenegro started in the 1990s, through research projects of the International Scientific Center for Ecology and Human Health MENEKO (Podgorica) [6], and resulted in a creation of radioecological map of Montenegro. *In situ* gamma spectrometry with HPGe detector [7], [8], had been applied for gamma radiation measurements across the country, on 70 points in total (including 14 beaches on the Adriatic Sea Coast). In such a way, activity concentrations of ^{238}U, ^{232}Th, ^{40}K and artificial ^{137}Cs had been determined in soils at the entire territory. Further analyses of radionuclide concentrations in soils of Montenegro were mainly based on standard procedure for soil sampling [9] and measurements by the method of laboratory gamma spectrometry using HPGe and NaI(Tl), but also the NaI(Tl) multidetector system [10], [11]. Based on the data related to ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soils, radiological impacts were considered for Montenegro [12] and some its areas, such as Mojkovac [13] and Nikšić [14] – where soil has also been analyzed for heavy metals and ^{238}U at a number of locations [15]. In addition to ^{137}Cs, plutonium isotopes ^{238}Pu and $^{239+240}$Pu were also considered for soils in Montenegro [16], [17].

* antovicn@yahoo.com

In spite of comprehensive research on natural radionuclides in Montenegro soils and beach sands, correlations among them have not been considered so far, excepting an evaluation of radioactivity in soil using statistical approach with a brief view on correlations which included ^{137}Cs as well [18].

The radionuclides of the ^{238}U and ^{232}Th series in rocks are in secular equilibrium, but in soils equilibrium can be disrupted since the radionuclides in the series show a wide range of geochemical behavior [19]. Correlations between terrestrial radionuclides in soils can be used to assess if they have the same geological origin and similar geochemical behavior, or for distinguishing natural background radiation and potential contamination from human activities. Some previous research showed that ratios of ^{238}U, ^{232}Th and ^{40}K activity concentrations can be a good monitoring indicator [20]. Significant positive correlation between activity concentrations of ^{232}Th and ^{238}U in soil has been observed in the other studies, for example in Serbia [21] and Slovenia – where positive correlation was also found between ^{226}Ra and ^{238}U [22].

This paper is aimed to evaluate correlation between ^{238}U and ^{232}Th in soil at the entire territory of Montenegro (hereinafter – Montenegro soil), and territory of Nikšić (Nikšić soil) which is characterized by a complex geology [15] and somewhat higher *indoor* radon concentrations [23]. It is also aimed to evaluate correlations between activities of ^{238}U and ^{232}Th in beach sands along Montenegro Coastline. The analysis is performed in order to determine activity ratios of these radionuclides in soils and sands, but also to establish baselines for future research on classification of soils according to geological settings which is based on natural radionuclides activity concentration.

The absorbed dose rate in air at 1 m above the ground caused by ^{238}U and ^{232}Th is calculated using a standard approach, i.e. using their activity concentrations in soil/sand and the dose coefficients from the UNSCEAR 2000 report [1]. This reference height (1 m) for the uniform distribution of radionuclides in soil represents the standard exposure zone for a person, and it is often used in assessments of outdoor external exposure to terrestrial gamma radiation.

2. Research Area and Methods

In a creation of radioecological map of Montenegro, the whole territory had been covered with a basic grid – 42 rectangles with approximate dimensions 22 km × 18 km [6]. In each of the rectangles *in situ* gamma spectrometry measurement was carried out on one measuring point, representative for the rectangle area by geological and soil characteristics. The portable gamma spectrometer consisted of the HPGe detector (n-type, beryllium window, 100 cm^3 active volume), multichannel analyzer and laptop computer. It had been used for two measurements on each point during the acquisition time of 2 h (detector at 1 m above the ground – 1 h; and detector at a depth of 10 cm in uncultivated or 20 cm in cultivated soil – 1 h). The other details about applied methodology can be seen in ref. [7], together with summary results.

The present study is focused on ^{238}U and ^{232}Th activity concentration in soil at each particular site of 42 in total (Fig. 1). The results for uncultivated topsoil (5 cm) from 24 points (in blue in Fig. 1) obtained after standard sampling, preparation and measurements in laboratory by the coaxial HPGe detector (ORTEC, with relative efficiency 40%) [12], are also used – for comparison and additional analysis.

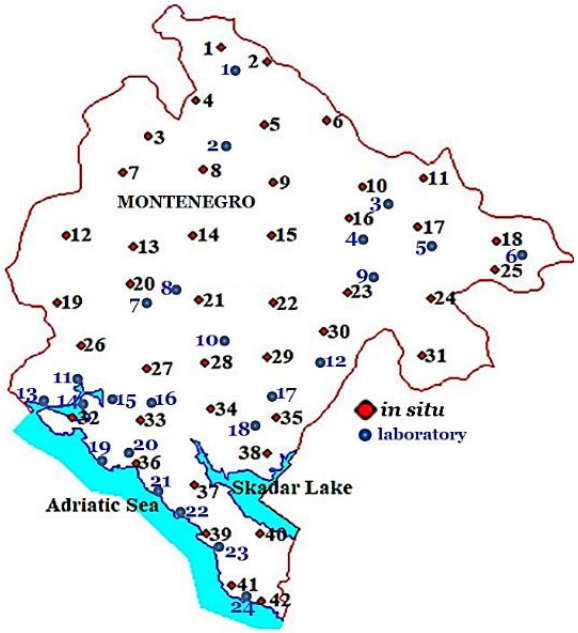

Figure 1. A map of Montenegro with soil points

The results of *in situ* [8] and laboratory [24] measurements of sands from renowned beaches along Coastline of Montenegro (Fig. 2) are re-used here in order to assess correlations among natural radionuclides, and evaluate associated dose rates.

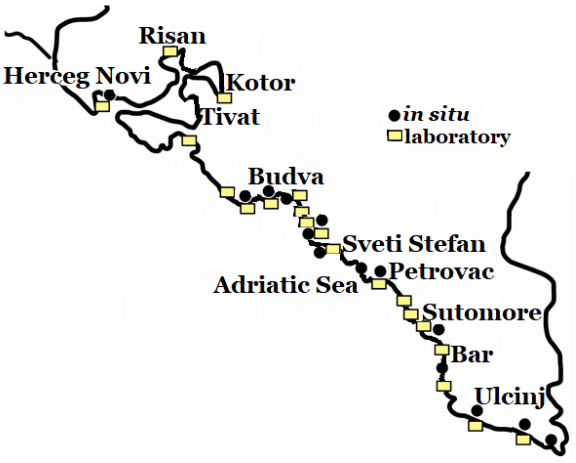

Figure 2. A map of Montenegro Coastline with sand points

It is important to note that a terrestrial (sand or soil) sample was taken from one point on a locality – for further preparation and measurement in laboratory, while in the case of *in situ* gamma spectrometry – obtained results are actually average values of radionuclide activities in sand/soil from an area around the detector, within a radius of 15 m.

The analysis of the correlations between ^{238}U and ^{232}Th activity in the Nikšić soil is based on the results of

laboratory HPGe spectrometry of surface soil from 12 locations shown in Fig. 3. Surface soil from 10 points in Nikšić had been previously analyzed for radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K [14], and these data for ^{232}Th are used in the present study, together with data for ^{238}U in the same soil samples. In order to cover typical geology of the Nikšić region, two additional soil points were chosen, uncultivated surface soil was sampled, prepared and measured (in 1L Marinelli beakers) in the standard procedures for the HPGe spectrometry (ORTEC, with relative efficiency 35%). Uranium-238 activity concentration in soil samples was determined using gamma spectra and the 1001 keV photopeak (gamma rays from the decay of ^{234m}Pa, i.e. de-excitation of ^{234}U). This high-energy photopeak is often used to quantify ^{238}U in soils and geological samples, as less affected by self-absorption and sample density in compare to the peaks at lower energies. The gamma rays from alpha decay of ^{238}U to ^{234}Th have very low intensities, as well as gamma rays from beta minus decay of ^{234}Th to ^{234m}Pa, excepting 63.3 keV (3.75%) [25] which suffers from significant self-absorption. The ^{232}Th/^{228}Ac activity concentration was determined using the photopeaks created by gamma rays 338 keV and 911 keV, the ^{226}Ra activity concentration – using the 295 keV, 352 keV, 609 keV, 1120 keV and 1764 keV photopeaks (gamma rays from beta minus decays of ^{214}Pb to ^{214}Bi and ^{214}Bi to ^{214}Po), and ^{40}K – using the photopeak created by the 1461 keV gamma rays. These intense photopeaks are used to calculate activity concentrations of radionuclides in the standard procedure (based on the net count rate under the photopeak, divided by the product of relative intensity of gamma ray, photopeak efficiency and mass of the dried sample).

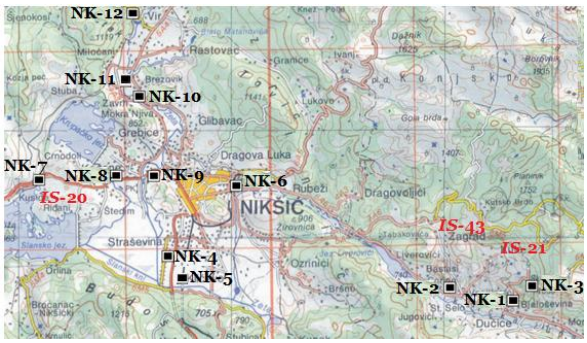

Figure 3. A map of soil points in the Nikšić region, with *in situ* points *IS*-20 and *IS* -21 (from basic grid in Fig. 1) and additional *in situ* point *IS*-43 (with geological base: 1 – Permo-Triassic clastic sediments, 3 – andesite volcanic and volcano-clastic rocks, 7 – Upper Cretaceous bedded limestone, 9 – Lower Cretaceous limestone, 12 – Middle Triassic dolomite-limestones, 2, 4-6, 8, 10-11 – Quaternary alluvial and fluvioglacial sediments, *IS*-43 – Upper Jurassic red bauxite deposits)

The absorbed dose rate is evaluated by multiplying activity concentration with corresponding dose coefficient – 0.462 nGy/h per Bq/kg for ^{238}U and 0.604 nGy/h per Bq/kg for ^{232}Th [1].

The *SPSS Statistics* (v. 20) and Microsoft Excel are used for statistical analysis. Descriptive statistics are used to summarize the key features of datasets. Quantile-Quantile (Q-Q) plots (plotting observed data against theoretical quantiles), Kolmogorov-Smirnov and Shapiro-Wilk tests are used to assess if datasets follow the normal distribution, which is assumed for radionuclide activity in soil when concentrations are expected to be relatively homogeneous (resulting from long-term geological processes). Correlations between ^{238}U and ^{232}Th activity concentrations in soil are considered through Pearson linear correlation coefficients, but also nonparametric Spearman monotonic correlation coefficients showing whether ^{238}U and ^{232}Th activity concentrations in soil increase together, not necessarily linearly. Additionally, linear regression is used to quantify and model the relationship between radionuclide activity concentrations for their statistically significant linear correlations.

3. Results and Discussion

The activity concentrations of ^{238}U and ^{232}Th at 42 points of Montenegro soil (MNE) are given in Table 1, while those for the Nikšić locations (NK) – in Table 2.

Table 1. U-238 and Th-232 in Montenegro soil (based on data from ref. [6])

Measuring site	^{238}U, Bq/kg	^{232}Th, Bq/kg
MNE-1	9.2 ± 1.1	26 ± 3
MNE-2	7.8 ± 1.1	25 ± 3
MNE-3	12 ± 1	16 ± 2
MNE-4	14 ± 2	14 ± 2
MNE-5	13 ± 1	20 ± 2
MNE-6	16 ± 2	16 ± 2
MNE-7	24 ± 3	31 ± 3
MNE-8	17 ± 2	19 ± 2
MNE-9	17 ± 2	23 ± 2
MNE-10	23 ± 2	27 ± 3
MNE-11	15 ± 2	14 ± 2
MNE-12	56 ± 4	33 ± 4
MNE-13	15 ± 2	21 ± 2
MNE-14	18 ± 2	23 ± 2
MNE-15	16 ± 2	19 ± 2
MNE-16	22 ± 2	21 ± 2
MNE-17	23 ± 2	28 ± 3
MNE-18	15 ± 2	17 ± 2
MNE-19	16 ± 2	22 ± 2
MNE-20	67 ± 7	44 ± 4
MNE-21	22 ± 2	17 ± 2
MNE-22	11.1 ± 1.1	14 ± 2
MNE-23	7 ± 0.7	9.3 ± 1.0
MNE-24	20 ± 2	25 ± 3
MNE-25	14 ± 2	14 ± 2
MNE-26	133 ± 8	26 ± 3
MNE-27	70 ± 7	37 ± 4
MNE-28	28 ± 3	28 ± 2
MNE-29	23 ± 2	16 ± 2
MNE-30	11.1 ± 1.5	16 ± 2
MNE-31	18 ± 2	18 ± 2
MNE-32	166 ± 8	74 ± 7
MNE-33	22 ± 2	20 ± 2
MNE-34	44 ± 4	27 ± 2
MNE-35	67 ± 7	41 ± 4
MNE-36	22 ± 2	26 ± 3
MNE-37	12 ± 1	17 ± 2
MNE-38	41 ± 4	37 ± 4
MNE-39	22 ± 2	20 ± 2
MNE-40	14 ± 2	17 ± 2
MNE-41	24 ± 2	20 ± 2
MNE-42	23 ± 3	18 ± 2

An average value of the ^{238}U activity concentrations in Montenegro soil (29.3 Bq/kg) is slightly below 35 Bq/kg which is median of mean values for soils worldwide [1]. An average ^{232}Th activity concentration (23.7 Bq/kg) is also below median of mean values for

soils worldwide (30 Bq/kg) [1]. At the same time, average values of the ^{238}U and ^{232}Th activity concentrations in the Nikšić soil (59.5 and 54.4 Bq/kg, respectively) exceed median of mean values for soils worldwide.

Table 2. U-238 and Th-232 in Nikšić soil

Location	^{238}U, Bq/kg	^{232}Th, Bq/kg
NK-1	70.9 ± 14.2	44.3 ± 1.6
NK-2	39.7 ± 13.9	45.4 ± 1.7
NK-3	72.5 ± 15.5	51.6 ± 1.8
NK-4	53.0 ± 13.3	33.9 ± 1.3
NK-5	43.6 ± 21.3	51.8 ± 2.0
NK-6	112 ± 16	46.9 ± 1.7
NK-7	55.8 ± 20.8	74.3 ± 2.6
NK-8	62.3 ± 17.3	53.2 ± 1.9
NK-9	77.4 ± 17	66.3 ± 2.3
NK-10	42.9 ± 14.6	51.1 ± 1.8
NK-11	40.3 ± 15.5	56.0 ± 2.0
NK-12	43.1 ± 13.4	78.6 ± 2.7

Statistical analysis showed that distributions of ^{238}U and ^{232}Th activity concentrations in soil deviate from the normal distribution. As an illustration, the normal Q-Q plot of the ^{232}Th concentrations in Montenegro soil is shown in Fig. 4, whereas results of the tests of normality are given in Table 3. Table 4 presents basic statistics, including the values of skewness and kurtosis.

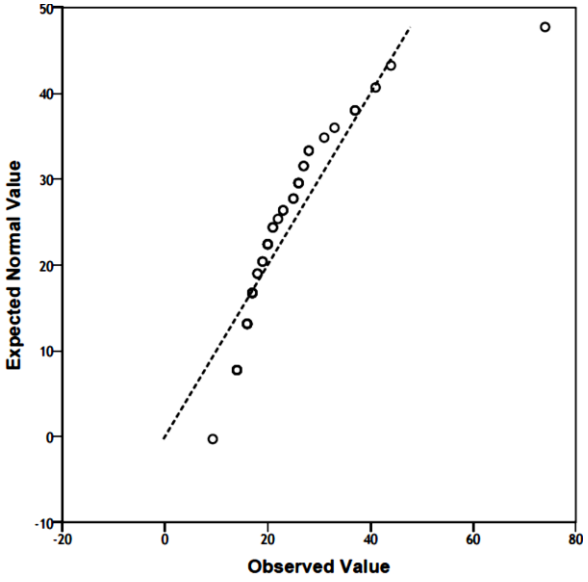

Figure 4. Normal Q-Q plot of the ^{232}Th concentrations in Montenegro soil

Table 3. U-238 and Th-232 in soil: tests of normality

	Kolmogorov-Smirnov[a] (KS)		Shapiro-Wilk (SW)	
	Statistic	*p*	Statistic	*p*
Montenegro				
^{238}U	0.352	0.000	0.588	0.000
^{232}Th	0.182	0.001	0.772	0.000
Nikšić				
^{238}U	0.188	0.200*	0.848	0.034
^{232}Th	0.205	0.173	0.927	0.353

[a] Lilliefors Significance Correction
*It is a lower bound of the true significance

Table 4. U-238 and Th-232 in soil: descriptive statistics

	U-238, Bq/kg (Montenegro)	U-238, Bq/kg (Nikšić)
Mean	29.3	59.5
Std. error of mean	4.87	6.15
Min-Max	7-166	39.7-112
Median	19.0	54.4
Std. deviation	31.6	21.3
Variance	997	454
Skewness	3.073	1.426
Kurtosis	10.235	2.280
	Th-232, Bq/kg (Montenegro)	Th-232, Bq/kg (Nikšić)
Mean	23.7	54.4
Std. error of mean	1.70	3.71
Min-Max	9.3-74	33.9-78.6
Median	20.5	51.7
Std. deviation	11.0	12.8
Variance	122	165
Skewness	2.593	0.650
Kurtosis	9.752	0.114

The highest ^{238}U concentration was found at the Montenegro *in situ* point 32 (Krašići – Tivat, in the Coastal region [7]), while the highest ^{232}Th concentration – at the same *in situ* point and the NK-12 (with the Middle Triassic dolomite-limestones in the geological base).

It is important to mention that part of the Nikšić region – Nikšićka Župa (Zagrad and Kuta – in Fig. 3), not far from NK-1 and NK-3, is characterized with Upper Jurassic red bauxite deposits. *In situ* measurement at the location Zagrad (*IS*-43) previously showed ^{238}U concentration of (70 ± 7) Bq/kg, and ^{232}Th – (137 ± 11) Bq/kg [6]. A comparison of these results with ones obtained by the laboratory gamma spectrometry for soils NK-1 and NK-3 (Table 2) shows that results for ^{238}U are comparable, whilst those for ^{232}Th differ significantly. A better agreement for ^{238}U in compare to that for ^{232}Th has also been found for the close points *IS*-20 (MNE-20 in Table 1) and NK-7. These differences are likely caused by different methodologies (*in situ* and laboratory measurements), i.e. different sampling approaches.

The ratio of ^{232}Th and ^{238}U activity concentrations in Montenegro soil ranges from 0.2 (MNE-26) to 3.2 (MNE-2), with an average of 1.1, while in the Nikšić soil – from 0.4 (NK-6) to 1.8 (NK-12), with an average of around 1. Taking the worldwide population-weighted average activity concentrations of ^{232}Th and ^{238}U in soils (45 and 33 Bq/kg, respectively – in the UNSCEAR 2000 [1] and UNSCEAR 2008 [2], 42 and 30 Bq/kg, respectively – in the UNSCEAR 2024 [19]), the ratio at the global level is 1.4, but it varies in different regions due to geological variability and environmental factors.

Correlation coefficients between ^{232}Th and ^{238}U activity concentrations in Montenegro and Nikšić soil are presented in Table 5. Statistically significant (at the 0.01 level) positive correlation is found for Montenegro soil (r = 0.805, r_S = 0.719), but not for the Nikšić soil.

Although further research is needed (including additional *in situ* points to obtain sufficiently dense network, particularly in some rectangles of the basic map), strong positive linear correlation between ^{232}Th and ^{238}U obtained in the present study indicates that their concentrations in soil are mostly influenced by common origin and similar geochemical behavior. A weak negative correlation between them in the Nikšić soil suggests dissimilar source/geochemical behavior or

alteration by distinct processes acting on the soil. It should be clarified in further research. For example, investigation of geochemical distributions can reveal influence of geological processes and lithological composition on terrestrial radionuclides transport and accumulation [26].

Table 5. Correlations of U-238 and Th-232 in soil (r: Pearson, r_S: Spearman's rho)

		^{238}U	^{232}Th
Montenegro			
^{238}U	*r*	1	
	rs	1.000	
^{232}Th	*r*	**0.805**	1
	rs	**0.719**	1.000
Nikšić			
^{238}U	*r*	1	
	rs	1.000	
^{232}Th	*r*	−0.143	1
	rs	−0.035	1.000

Since statistically significant correlation between ^{238}U and ^{232}Th activity in the Nikšić soil was not found, correlations among them and the other natural radionuclides (such as ^{226}Ra and ^{40}K) were then studied, using the data from ref. [14] (^{232}Th, ^{226}Ra and ^{40}K) and measuring results for two additional soil points. For the NK-5, ^{226}Ra and ^{40}K are found to be (48.2 ± 1.7) Bq/kg and (480 ± 16) Bq/kg, respectively, while for the NK-11 – (44.8 ± 1.52) Bq/kg and (284 ± 10) Bq/kg, respectively. Significant (at the 0.01 level) correlation is found for ^{232}Th and ^{226}Ra in the Nikšić soil (r = 0.856, rs = 0.776), and not for ^{238}U and ^{226}Ra, ^{238}U and ^{40}K, ^{226}Ra and ^{40}K, or ^{232}Th and ^{40}K.

A strong correlation between ^{232}Th and ^{226}Ra is also found for the Montenegro soil measured in laboratory. Namely, statistical analysis related to soils from 24 points (blue in Fig. 1), having ^{232}Th concentrations from 18.2 to 107 Bq/kg (43.6 Bq/kg in average) and ^{226}Ra – from 11.0 to 216 Bq/kg (39.9 Bq/kg in average) [12], showed correlation coefficients r = 0.893 and rs = 0.826. The highest activity concentrations were again determined for the Tivat region (Plavi Horizonti, i.e. blue point 14 in Fig. 1). This analysis confirmed a deviation from the normal distribution. The KS and SW tests gave statistic/p value of 0.168/0.079 and 0.854/0.003, respectively (^{232}Th), as well as 0.341/0.000 and 0.535/0.000, respectively (^{226}Ra).

The strong positive linear correlations ^{232}Th and ^{238}U and ^{232}Th and ^{226}Ra for Montenegro soil, as well as ^{232}Th and ^{226}Ra for the Nikšić soil, suggest a possibility to predict the level of one radionuclide based on the activity concentration of the other one. In order to examine such a possibility, linear regression is performed for the revealed significant correlations and results are shown in Fig. 5 (where A_{Th}, A_U and A_{Ra} are activity concentrations of ^{232}Th, ^{238}U and ^{226}Ra, respectively). Statistically significant prediction (p < 0.05) of the ^{232}Th activity concentration based on the value of ^{238}U is found for Montenegro soil, which is shown in Fig. 5a together with the regression equation and R square (indicating how much of the variation in the dependent variable can be explained by the predictor). As can be seen from Fig. 5a, 64.8% of the variation in ^{232}Th activity could be explained by ^{238}U as a predictor. A larger variation in the ^{232}Th activity concentration can be explained by ^{226}Ra as a predictor – 73.2% in the Nikšić soil (Fig. 5b), and 80.8% in the Montenegro soil analyzed in laboratory (Fig. 5c).

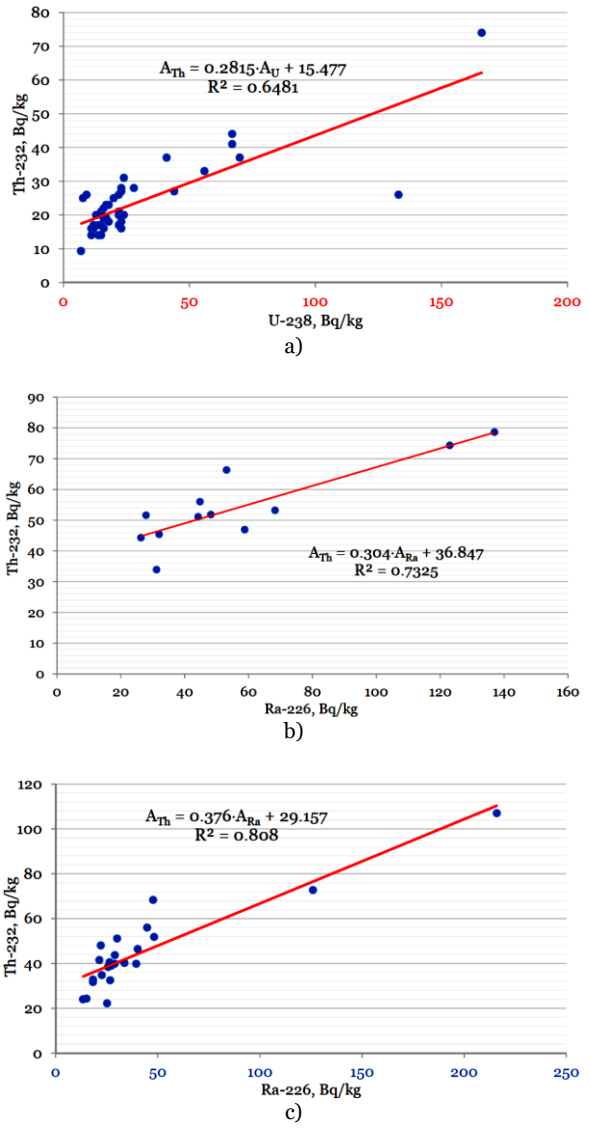

Figure 5. Linear regression for statistically significant linear correlations: Th-U for Montenegro soil (a), Th-Ra for Nikšić soil (b), Th-Ra for Montenegro soil (c)

The absorbed dose rates in air from ^{238}U in soil (D_U, calculated as the product of A_U and 0.462 nGy/h per Bq/kg [1]) and ^{232}Th in soil (D_{Th}, calculated as the product of A_{Th} and 0.604 nGy/h per Bq/kg [1]) are shown in Table 6 and Fig. 6. For a comparison, the global medians of absorbed dose rates in air calculated from medians of average concentrations of ^{238}U and ^{232}Th in soils worldwide were found to be 16 nGy/h – from ^{238}U in soil, and 18 nGy/h – from ^{232}Th in soil [1]. The global average of the population-weighted absorbed dose rate in air from terrestrial gamma radiation outdoors has been estimated at 59 nGy/h [1]. However, the newest estimate given by the UNSCEAR shows lower global average of the population-weighted absorbed dose rate in air (49 nGy/h, with an estimated uncertainty of around 10% [19]).

The absorbed dose rates presented here are basis for evaluation of external exposure outdoors, since corresponding annual effective dose is the product of

the absorbed dose rate in air (calculated or directly measured), the outdoor occupancy factor (0.2), the dose coefficient converting absorbed dose in air to effective dose (0.7 Sv/Gy, if it is the effective dose received by adults), and 8760 h (the number of hours in a year) [1]. At the same time, activity concentrations of terrestrial radionuclides in soil are basis not only for future research on their geochemical behavior, but also for evaluation of the soil-plant transfer factors, since the soil–plant–man chain has been recognized as one of the major pathways for transfer of radionuclides to human body [3].

Table 6. Dose rates from U-238 and Th-232 in soil

	D_U, nGy/h (Montenegro)	D_U, nGy/h (Nikšić)
Mean	13.5	27.5
Std. error of mean	2.25	2.84
Min-Max	3.23-76.7	18.3-51.7
Median	8.78	25.1
Std. deviation	14.6	9.85
	D_{Th}, nGy/h (Montenegro)	D_{Th}, nGy/h (Nikšić)
Mean	14.3	32.9
Std. error of mean	1.03	2.24
Min-Max	5.62-44.7	20.5-47.5
Median	12.4	31.2
Std. deviation	6.67	7.75

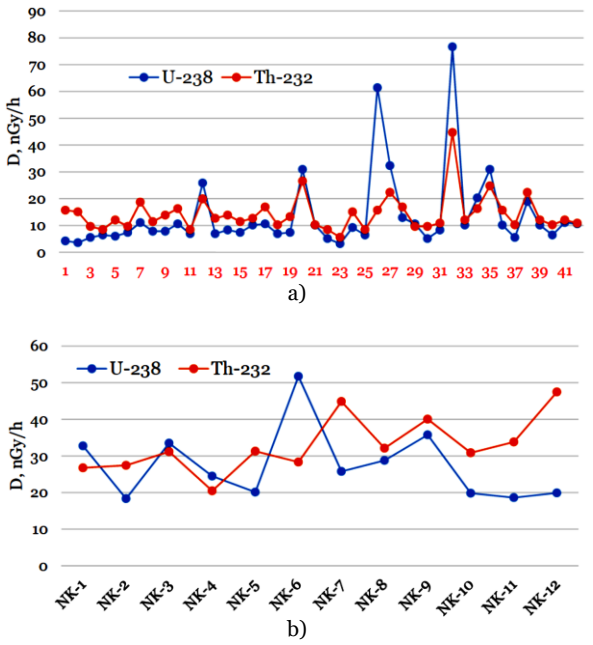

Figure 6. Absorbed dose rate in air from ^{238}U and ^{232}Th in soil: Montenegro (a), Nikšić (b)

Uranium-238 and ^{232}Th activity concentrations in sands on the 14 beaches (*in situ* in Fig. 2), from the Herceg Novi (Igalo) to Ulcinj (Ada Bojana), can be seen in Fig. 7. The range, arithmetic mean and standard deviation were found to be 2-16, 7.34 and 3.41 Bq/kg, respectively (^{238}U), and 0.8-11.8, 4.14 and 3.57 Bq/kg, respectively (^{232}Th) [8]. The ratio of ^{232}Th and ^{238}U activity concentrations in these beach sands ranged from 0.08 to 1.18, with an average of 0.55. The absorbed dose rates in air caused by ^{238}U in beach sands ranged from 0.92 to 7.39 nGy/h, with an average and standard deviation of 3.39 and 1.57 nGy/h, respectively. The doses in air coming from ^{232}Th in beach sands ranged from 0.48 to 7.13 nGy/h, with an average and standard deviation of 2.50 and 2.16 nGy/h, respectively.

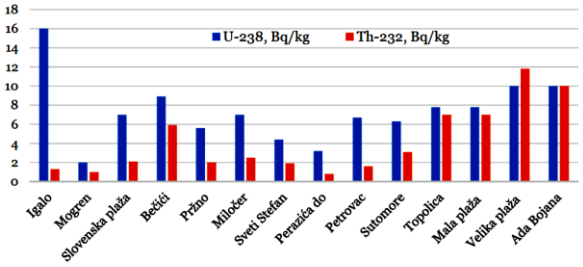

Figure 7. U-238 and Th-232 in beach sands (based on data from ref. [8])

The normal Q-Q plots and tests of normality showed that distribution of ^{232}Th activity concentration in beach sands deviate from normal distribution more than the ^{238}U distribution. The KS and SW tests gave statistic/*p* value of 0.160/0.200 and 0.929/0.293, respectively (^{238}U), as well as 0.258/0.012 and 0.830/0.012, respectively (^{232}Th).

The correlation analysis showed that there is no statistically significant linear correlation between ^{238}U and ^{232}Th activity concentrations in beach sands (r = 0.390, *Sig*. 0.169), although there is a significant (at the 0.05 level) monotonic correlation (r_S = 0.640, *Sig*. 0.014). Therefore, correlations among them and ^{40}K activity concentrations in beach sands (from 16 to 236 Bq/kg, and 87.2 Bq/kg in average [8]) are also analyzed. Significant linear correlation between ^{238}U and ^{40}K was not found (r = 0.330, *Sig*. 0.249), while for ^{232}Th and ^{40}K it is very strong statistically significant positive correlation – linear (r = 0.939, *Sig*. 0.000) and monotonic (r_S = 0.963, *Sig*. 0.000).

The regression analysis confirmed linear correlation between ^{232}Th and ^{40}K in beach sands, with R^2 = 0.8814 and regression equations: $A_{Th} = 0.044 \cdot A_K + 0.3084$ and $A_K = 20.047 \cdot A_{Th} + 4.1623$. Positive correlation between ^{40}K and ^{232}Th was observed for example in soils of Serbia [21] and Slovenia [22], but also in some beach sediments in India [27].

In order to check the correlation between ^{232}Th and ^{40}K in beach sands, additional analysis was performed with data related to the 20 beaches along the Coastline of Montenegro (yellow rectangles in Fig. 2, from Herceg Novi – Njivice, to Ulcinj – Velika plaža), obtained by the laboratory HPGe spectrometry [24]. Radium-226 activity concentration (Fig. 8) in these beach sands ranged from 2.09 to 15.5 Bq/kg with an average of 7.41 Bq/kg, whereas ^{232}Th – from 1.37 to 16.6 Bq/kg (5.16 Bq/kg in average) and ^{40}K – from 7.13 to 305 Bq/kg, (97.3 Bq/kg in average) [24]. These ^{232}Th activities give the absorbed dose rate in air at 1 m above sand – from 0.83 to 10.0 nGy/h (mean: 3.12 nGy/h), as ^{226}Ra activities give dose rates from 0.97 to 7.16 nGy/h (mean: 3.42 nGy/h).

The analysis confirmed very strong positive correlation between ^{232}Th and ^{40}K in beach sands (r = 0.933, r_S = 0.874), and linear regression analysis shows R^2 = 0.8713 and equations: $A_{Th} = 0.0425 \cdot A_K + 1.0282$ and $A_K = 20.512 \cdot A_{Th} - 8.5633$.

For a comparison, results for the same beach are shown in Fig. 9, indicating that *in situ* and laboratory gamma spectrometry gave different results for example

in the case of Bečići, and very similar results for ^{232}Th and ^{40}K in sand at the Mogren or Miločer beach.

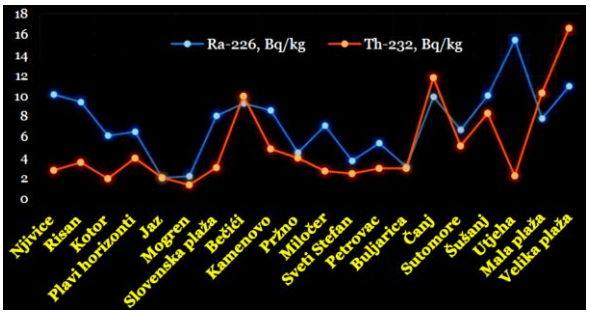

Figure 8. Ra-226 and Th-232 in beach sands (based on data from ref. [24])

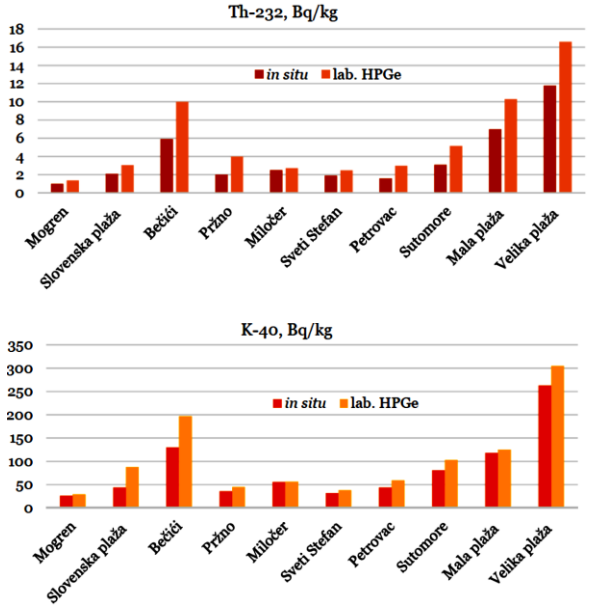

Figure 9. Th-232 and K-40 in beach sands (based on data from refs. [8] and [24])

4. Summary and Perspectives

The ^{232}Th and ^{238}U activity concentrations are found to be significantly positively correlated in soil at the territory of Montenegro as a whole, but not in soil of Nikšić or in sand at beaches on the Montenegrin Coast. The results presented in this paper are baselines for future research on radionuclides origin and geochemical behavior, as well as for classification of soils in Montenegro according to geological settings – based on natural radionuclides activity concentration.

Further research should include additional measuring points, soil properties and geological characteristics (geotectonic units, geological formations and subformations), as well as multivariate statistical techniques, such as cluster analysis which can be used to map geochemical variations. Additional statistical analysis should involve tools able to separate geological formations or soil types by radionuclide activity concentrations (such as Discriminant Analysis), as well as tools summarizing patterns among radionuclide activity concentrations (such as Principal Component Analysis as the extraction method in the factor analysis).

An average absorbed dose rate in air coming from ^{238}U in Montenegro soil is found to be slightly lower than the global average, as well as the dose coming from ^{232}Th. In opposite, both dose rates (from ^{238}U and from ^{232}Th) for the Nikšić region are found to be higher than the global averages. In regard to the dose rates from ^{238}U and ^{232}Th in beach sands – they do not exceed 7.4 nGy/h (^{238}U) and 10 nGy/h (^{232}Th).

The absorbed dose rates obtained in this study are valuable for the mapping, i.e. creating a complete picture of external exposure outdoors in Montenegro. Furthermore, the results are the basis for planning a network of new points where measurements should be conducted using laboratory techniques or *in situ* methodology – to ensure a sufficiently dense network covering soils, beach sands and sediments.

References

1. *Sources and effects of ionizing radiation*, Annex B, UNSCEAR 2000 Report to the General Assembly with Scientific Annexes, UNSCEAR, New York (NY), USA, 2000.
2. *Sources and effects of ionizing radiation*, Annex B, UNSCEAR 2008 Report to the General Assembly with Scientific Annexes, UNSCEAR, New York (NY), USA, 2010.
3. V. A. Pulhani, S. Dafauti, A. G. Hegde, R. M. Sharma, U. C. Mishra, "Uptake and distribution of natural radioactivity in wheat plants from soil," *J. Environ. Radioact.*, vol. 79, no. 3, pp. 331 – 346, Nov. 2005. DOI: 10.1016/j.jenvrad.2004.08.007 PMid: 15607519
4. M. Brai et al., "Correlation of radioactivity measurements, air kerma rates and geological features of Sicily," *Radiat. Meas.*, vol. 41, no. 4, pp. 461 – 470, Apr. 2006. DOI: 10.1016/j.radmeas.2005.09.004
5. A. Navas, J. Soto, J. Machin, "238U, 226Ra, 210Pb, 232Th and 40K activities in soil profiles of the Flysch sector (Central Spanish Pyrenees)," *Appl. Radiat. Isot.*, vol. 57, no. 4, pp. 579 – 589, Oct. 2002. DOI: 10.1016/S0969-8043(02)00131-8 PMid: 12361338
6. П. Вукотић и др., *Фон гама-зрачења у Црној Гори (1993-1995)*, Међународни научни центар за екологију и здравље човјека МЕНЕКО, Подгорица, Црна Гора, 1996. (P. Vukotić et al., *Background gamma-radiation in Montenegro (1993-1995)*, International Scientific Center for Ecology and Human Health MENEKO, Podgorica, Montenegro, 1996.)
7. P. Vukotić et al., "Background gamma-radiation in Montenegro," in *Proc. IRPA Reg. Symp. Radat. Prot. in Neighbouring Countries of Central Europe*, Prague, Czech Republic, Sep. 1997, pp. 477 – 479.
8. P. Vukotić et al., "Radioactivity on the Montenegrin coast, Yugoslavia," *J. Radioanal. Nucl. Chem.*, vol. 235, no. 1 – 2, pp. 151 – 157, Sep. 1998. DOI: 10.1007/BF02385954
9. N. A. Chieco, *The Procedures Manual of the Environmental Measurements Laboratory*, 28th ed., U.S. Department of Energy, New York (NY), USA, 1997.
10. N. M. Antovic, N. Svrkota, I. Antovic, "Measuring 226Ra and 232Th activity in soil and vegetation samples using a method of double γ-coincidences," *J. Radioanal. Nucl. Chem.*, vol. 283, no. 2, pp. 313 – 318, Feb. 2010. DOI: 10.1007/s10967-009-0376-0
11. N. M. Antovic, N. Svrkota, I. Antovic, V. Popovic, P. Vukotic, "Measuring 137Cs, 40K and decay products of 226Ra and 232Th in samples of different

nature by a multidetector spectrometer," *J. Radioanal. Nucl. Chem.*, vol. 290, no. 1, pp. 81 – 88, Oct. 2011.
DOI: 10.1007/s10967-011-1136-5

12. N. M. Antovic, N. Svrkota, I. Antovic, "Radiological impacts of natural radioactivity from soil in Montenegro," *Radiat. Prot. Dosim.*, vol. 148, no. 3, pp. 310 – 317, Feb. 2012.
DOI: 10.1093/rpd/ncr087
PMid: 21498861
13. N. M. Antović, N. R. Svrkota, D. S. Bošković, I. M. Antović, "Radioactivity in soil from Mojkovac, Montenegro, and assessment of radiological and cancer risk," *Nucl. Technol. Radiat.*, vol. 27, no. 1, pp. 57 – 63, Mar. 2012.
DOI: 10.2298/NTRP1201057A
14. N. M. Antović et al., "Radioactivity impact assessment of Nikšić region in Montenegro," *J. Radioanal. Nucl. Chem.*, vol. 302, no. 2, pp. 831 – 836, Nov. 2014.
DOI: 10.1007/s10967-014-3254-3
15. Н. Свркота, Н. Антоновић, Р. Жижић, Р. Зекић, "238U и тешки метали у земљишту Никшића, Црна Гора," у *Зборник XXVIII симпозијума Друштва за заштиту од зрачења Србије и Црне Горе*, Вршац, Србија, 2015, стр. 148 – 154. (N. Svrkota, N. Antonovic, R. Zizic, R. Zekic, "238U and heavy metals in soils of Nikšić, Montenegro," in *Proc. XXVIII Symp. Soc. Radiat. Prot. Serbia and Montenegro*, Vršac, Serbia, 2015, pp. 148 – 154.)
16. N. M. Antovic, P. Vukotic, N. Svrkota, S. K. Andrukhovich, "First measurements of 238Pu and 238Pu/137Cs activity ratio in Montenegro soil," *J. Radioanal. Nucl. Chem.*, vol. 293, no. 1, pp. 299 – 302, Jul. 2012.
DOI: 10.1007/s10967-012-1661-x
17. N. M. Antovic, P. Vukotic, N. Svrkota, S. K. Andrukhovich, "Pu-239+240 and Cs-137 in Montenegro soil: their correlation and origin," *J. Environ. Radioact.*, vol. 110, pp. 90 – 97, Aug. 2012.
DOI: 10.1016/j.jenvrad.2012.02.001
PMid: 22445877
18. N. M. Antović, N. R. Svrkota, "Evaluation of radioactivity in Montenegro soil using a statistical approach," in *Proc. Int. Conf. Radiation Applications (RAP 2019)*, Belgrade, Serbia, Sept. 2019, pp. 90 – 95.
DOI: 10.37392/RapProc.2019.18
19. *Sources, effects and risks of ionizing radiation*, Annex B, UNSCEAR 2024 Report to the General Assembly with Scientific Annexes, UNSCEAR, New York (NY), USA, 2025.
20. M. Krmar, E. Varga, J. Slivka, "Correlations of natural radionuclides in soil with those in sediment from the Danube and nearby irrigation channels," *J. Environ. Radioact.*, vol. 117, pp. 31 – 35, Mar. 2013.
DOI: 10.1016/j.jenvrad.2011.12.023
PMid: 22244685
21. S. Dragović et al., "Lithogenic radionuclides in surface soils of Serbia: Spatial distribution and relation to geological formations," *J. Geochem. Explor.*, vol. 142, pp. 4 – 10, Jul. 2014.
DOI: 10.1016/j.gexplo.2013.07.015
22. T. Kovács et al., "Systematic survey of natural radioactivity of soil in Slovenia," *J. Environ. Radioact.*, vol. 122, pp. 70 – 78, Aug. 2013.
DOI: 10.1016/j.jenvrad.2013.02.007
PMid: 23558252
23. N. Antovic, P. Vukotic, R. Zekic, R. Ilic, "Indoor radon concentrations in the town of Niksic, Montenegro," *Radiat. Prot. Dosim.*, vol. 124, no. 4, pp. 385 – 391, May 2007.
DOI: 10.1093/rpd/ncm225
PMid: 17681959
24. N. M. Antovic, N. Svrkota, I. Antovic, R. Svrkota, D. Jancic, "Radioactivity in Montenegro beach sands and assessment of the corresponding environmental risk," *Isot. Environ. Health Stud.*, vol. 49, no. 2, pp. 153 – 162, Jun. 2013.
DOI: 10.1080/10256016.2013.734303
PMid: 23452289
25. *Nuclear data/Tables of recommended data*, Laboratoire National Henri Becquerel, Buc, France, 2025.
Retrieved from: lnhb.fr
Retrieved on: Feb. 9, 2025
26. Q. Wang et al., "Geochemical distributions of natural radionuclides in surface soils and sediments impacted by lead-zinc mining activity," *Ecotoxicol. Environ. Saf.*, vol. 263, 115210, Sep. 2023.
DOI: 10.1016/j.ecoenv.2023.115210
PMid: 37418943
27. M. SureshGandhi et al., "Measurements of natural gamma radiation in beach sediments of north east coast of Tamilnadu, India by gamma ray spectrometry with multivariate statistical approach," *J. Radiat. Res. Appl. Sci.*, vol. 7, no. 1, pp. 7 – 17, Jan. 2014.
DOI: 10.1016/j.jrras.2013.11.001