ANALYSIS OF SPECIFIC RADIONUCLIDE ACTIVITY VARIATIONS IN SOIL WITHIN GEOTECTONIC UNITS OF REPUBLIC OF NORTH MACEDONIA

by

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To establish baseline values for concentrations of terrestrial radionuclides for the Republic of North Macedonia, a survey covering the entire territory was performed. The 213 soil samples were collected from regions around the major settlements and cities, approximately evenly distributed over the geotectonic units which constitute the country's geological foundation. The specific activities of radionuclides were measured by gamma spectrometry. The following geometric mean values and geometric standard deviations were obtained: 550 Bqkg^{-1} (1.47) for ^{40}K, 37 Bqkg^{-1} (1.53) for ^{226}Ra, 38 Bqkg^{-1} for ^{238}U, and 38 Bqkg^{-1} (1.53) for ^{232}Th in dry soil. The relation between specific activities of natural radionuclides and geology was investigated. Correlation between radionuclides, which may serve as additional geochemical indicators, and geology could not be found convincingly. Mutual correlations between ^{226}Ra, ^{238}U, and ^{232}Th activities were found to be high (Spearman ρ about 0.8), whereas the ones between these and ^{40}K are a bit lower, somewhat above ρ = 0.6.

Key words: ^{40}K, ^{226}Ra, ^{238}U, ^{232}Th, gamma spectrometry, soil, geology, lithostratigraphic unit

INTRODUCTION

Naturally occurring radionuclides: ^{40}K and radionuclides from ^{238}U and ^{232}Th chains in soil and rocks, as well as in the building materials, are the main sources of external exposure to humans. In addition, through their transfer into the food and water, they become potential sources for internal exposure through ingestion. Furthermore, the two radioactive noble gases ^{222}Rn and ^{220}Rn, which are members of the ^{238}U and ^{232}Th decay chains, which are present in the soil and building materials, emanating from the surface and are accumulated in the indoor environment, can be significant sources of internal exposure due to inhalation.

This has motivated surveys of terrestrial radiation all over the world. Among the purposes are the following:

establishing background levels, sometimes for evidence preservation; this way, a global radiation inventory is being built and quantities being mapped, see *e.g*. the European Atlas of Natural Radiation
(https://remon.jrc.ec.europa.eu/About/Atlas-of-Natural-Radiation),

radiation protection, including dose assessment,

resource exploration through surveys of geogenic radionuclides; the latter of the two activities aim to discover anomalies, to be defined against the background, and

scientific objectives, such as an investigation of relations with environmental control factors (*e.g*. geology), modelling of spatial variability, *etc*.

The global researches also include investigation of terrestrial radioactivity in different areas of the Balkan region. Some of the recent studies are generally related to establishing the background: radioactivity in soil, collected from the territories of: Serbia [1]; Western Serbia [2]; Serbian spas [3, 4]; Kastela Bay, Croatia [5]; Bosna and Herzegovina [6]; rural areas of Kosovo and Metohija [7]; Additional purposes are to discover anomalies of geogenic radionuclides in the

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surrounding of the largest coal-fired power plant in Serbia [8] and at the site of a closed Bulgarian uranium mine [9]. In the Republic of North Macedonia, an intensive campaign for the radioactive background determination began in 2008. Some results have already been published, such as the geographical variation of indoor radon and thoron concentrations [10-15], the relation between indoor radon and ^{226}Ra in soils [16], and the radioactivity in raw materials and end product from cement industry [17]. Furthermore, the spatial variations of radionuclides in soils in the area of two cities: Veles [18] and Kavadarci [19] were examined. In the study presented here, the spatial variability of radionuclides over the geotectonic units of North Macedonia in topsoil was investigated. Besides establishing representative baseline data for North Macedonia, the main goal of this study was to find the relation between the variability of the radionuclides' specific activity and one of geographical position and geological control factors.

MATERIALS AND METHODS

Study area, sample collection, and preparation

Geologically, the territory of the Republic of North Macedonia (25,713 km^2) includes various types of volcanic, sedimentary and metamorphic rocks of different age, genesis and mineral content. The territory can be divided into four geotectonic zones: Western zone (WZ), Pelagonijan Massif (PM), Vardar zone (VZ), and Serbo-Macedonian Massif (SMM) [20]. Between the Serbo-Macedonian Massif and the Vardar zones an area of young vulcanite is located, denoted as Kratovo-Zletovo area (KZA) [21].

A total number of 213 samples of undisturbed surface soil were collected from the locations in the vicinity of the settlements and cities shown in fig. 1, during the period of 2008-2010. Sampling was performed using a steel tool that samples exactly at the depth of 0-20 cm [22]. Most of the stones, pebbles and organic materials were removed from the samples. The samples were then dried, sieved and filled into standard Marinelli beakers of 500 cm^3. They were kept air tightly closed for one month in order to reach equilibrium between ^{226}Ra and its short-lived decay products. After this procedure, the samples were subjected to gamma spectrometry measurements.

Gamma spectrometry measurements

The specific activity of each of the investigated radionuclides was measured by gamma spectrometry. The activity of ^{40}K was determined from the 1460 keV line, whereas the activity of ^{226}Ra from the gamma lines of ^{214}Pb (295.22 keV, 351.93keV), ^{214}Bi (609.31 keV, 1120.29 keV, 1764.49keV); ^{238}U was determined from the gamma lines of ^{234}Th (63.28 keV) and ^{234}mPa (1001.03 keV) and finally, the activity of ^{232}Th was determined from the gamma lines of ^{228}Ac (338.32 keV, 911.2 keV, 968.97 keV) and ^{208}Tl (583.19 keV) [23]. The analysis included subtraction of the background spectrum, correction for interfering

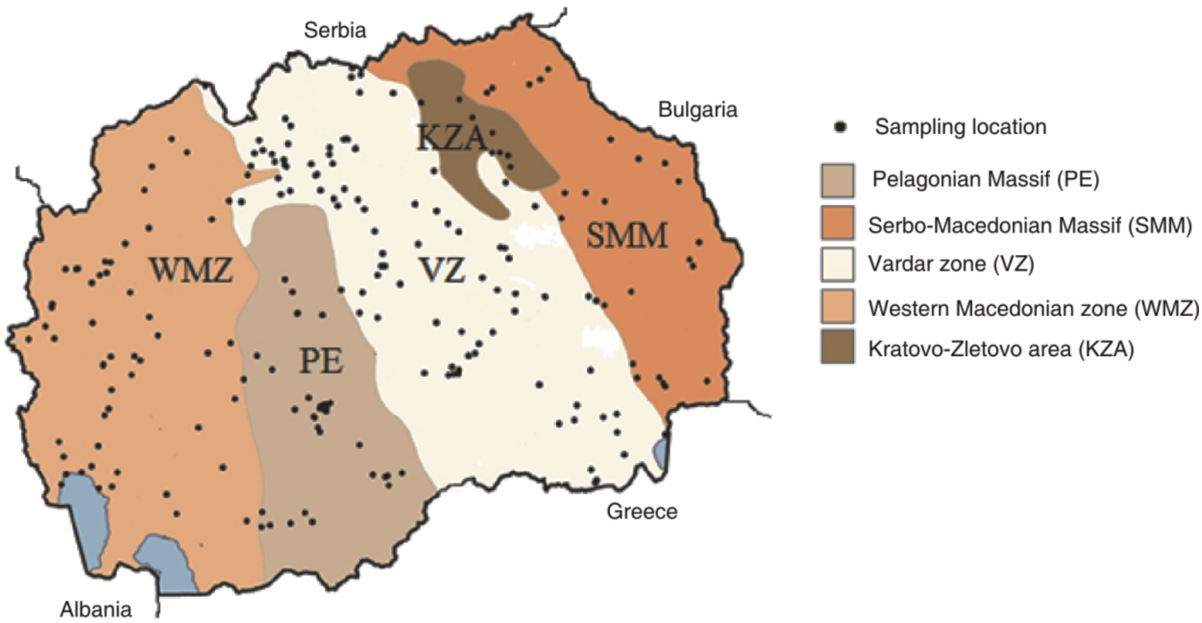

Figure 1. Spatial distribution of sampling locations over four geotechnical zones and one area of the Republic of North Macedonia

lines and correction for self-absorption. Quality assurance was performed with two reference materials from the International Atomic Energy Agency, IAEA-385 and PT IAEA-CU-2009-03 proficiency test. Measurement times were chosen such that the total relative combined uncertainty for each radionuclide was <5 %, at 68 % (1 sigma) confidence levels.

RESULTS AND DISCUSSION

Data characterization

Descriptive statistics of the ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities in soil samples are summarized in tab. 1. Kolmogorov-Smirnov distribution test confirmed the hypothesis that the samples that come from lognormally distributed populations, cannot be rejected at 95 % significance level, which is also visually suggested by the shapes of the histograms (fig. 2) for all the measured quantities. Their central tendency is represented by the geometric mean (GM) value, while the dispersion of the results is given as geometric standard deviation (GSD).

Table 1. Descriptive statistic of measured data

Statistic	*A* (dry) [Bqkg^{-1}]			
	^{40}K	^{226}Ra	^{238}U	^{232}Th
Number of observations	213	213	213	213
Minimum	80	9	9	7
Maximum	1390	123	111	145
Median	584	37	39	39
Arithmetic mean	585	41	41	41
Standard deviation	192	18	17	18
Geometric mean	550	37	38	38
Geometric standard deviation	1.47	1.53	1.53	1.54

Specific activities of natural radionuclides ranged from 80 to 1390 Bqkg^{-1} (GM: 550 Bqkg^{-1}) for ^{40}K, from 9 to 123 Bqkg^{-1} (GM: 37 Bqkg^{-1}) for ^{226}Ra, from 9 to 111 Bqkg^{-1} (GM: 38 Bqkg^{-1}) for ^{238}U and from 7 to 145 Bqkg^{-1} (GM: 38 Bqkg^{-1}) for ^{232}Th. From tab. 1 it is obvious that geographical variability, quantified by the geometric standard deviation, is quite uniform for all the radionuclides.

Generally, the GM values of the ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities were found to be slightly higher in comparison to the worldwide median activities of 400 Bqkg^{-1} for ^{40}K, 35 Bqkg^{-1} for ^{226}Ra,

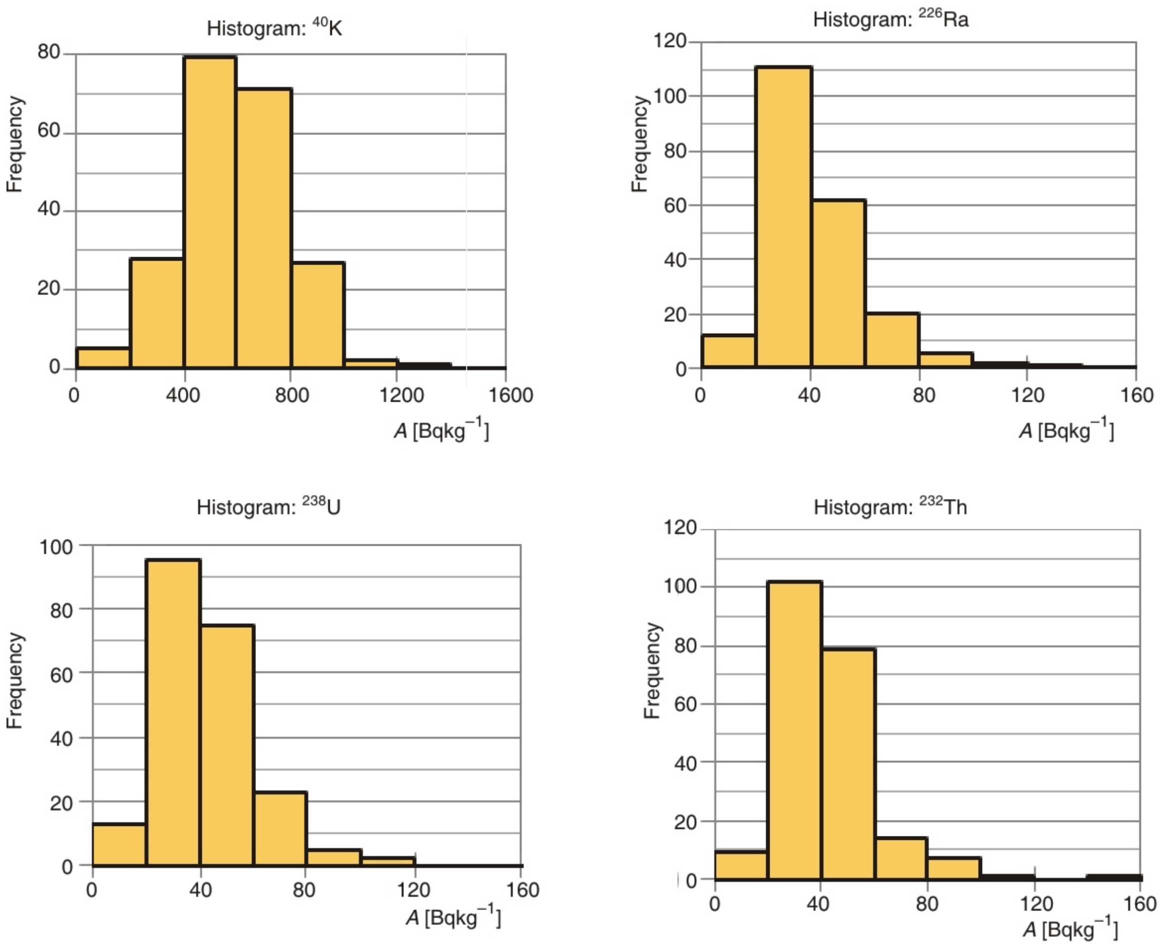

Figure 2. Histograms of the ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities

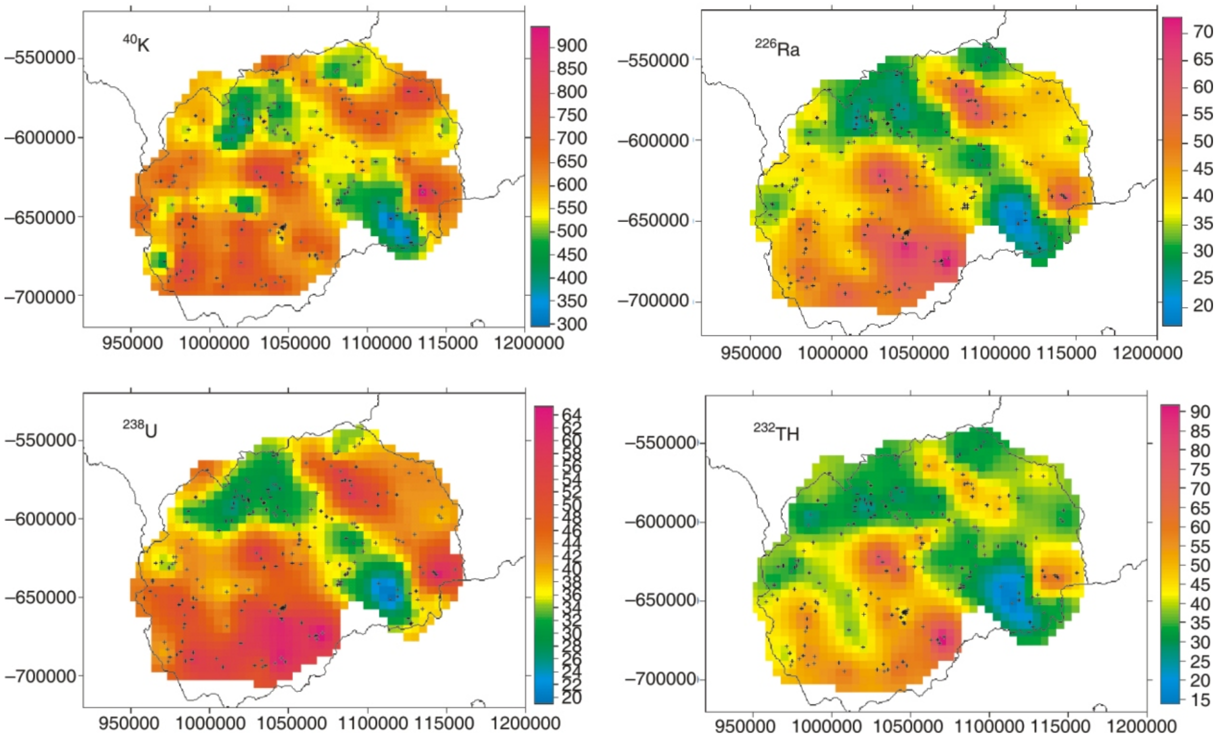

Figure 3. The maps of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities

35 Bqkg^{-1} for ^{238}U and 30 Bqkg^{-1} for ^{232}Th, given in UNSCEAR report [24]. They fall well in the ranges of the mean values reported for East and South European countries in the same report.

Specific activities spatial variation

Figure 3 presents the spatial distribution of measured specific activities for each radionuclide separately. The maps have been prepared by ordinary kriging based on variograms using Surfer 8 software. There are many published scientific papers from environmental science and radioecology in particular, which have introduced and discussed these methods. As a few examples among many others, most originating in radon studies, we cite [16, 25-27] for the application of ordinary kriging and variants. In [16, 28-30], different interpolation methods, including kriging, have been compared. Pasztor *et al*., used regression kriging for accounting for covariates [31], while in [16] this has been attempted by establishing a bivariate distribution model. Raspa *et al*., used disjunctive kriging for estimating local exceedance probability [32]. Spatial means are taken from the grid statistics and leave-one-out cross-validation correlations between observed and estimated concentrations can be found in tab. 2.

Comparison with tab. 1 shows that raw and modelled means are very similar. Raw means (tab. 1) may contain a bias because samples have not been taken spatially randomly or according to a grid, thus a preferential sampling effect may be present. On the other hand, the modelled means may be influenced by modelling errors (*e. g*., the variograms are built from the raw values and the fitted variogram models have uncertainty). Regarding cross-validation correlations, according to Cohen's rule of thumb, $r > 0.3$ can be considered medium, $r > 0.5$ large effects [33, 34].

Table 2. Spatial means of radionuclide concentrations (Bqkg^{-1} per dry) and cross-validation correlation coefficients

Radionuclide	AM	Median	XVal Pearson r
^{40}K	590	593	0.42
^{226}Ra	40.8	39.8	0.61
^{238}U	42.3	41.7	0.55
^{232}Th	41.7	39.7	0.66

It is obvious and no surprise, by what is known from geochemistry, that the specific activities of natural radionuclides are directly related to the geotectonic division of the country. The lower values were found along the central part of the territory (direction south of the north) which belongs to the Geotectonic Vardar zone, while the higher values were mostly found in the Pelagonia and Krusevsko Zletovska area.

Relations of specific activities with geology

Geological information was associated with each sampling location to investigate to which degree radionuclide concentrations are controlled by geol-

Table 3. Range, GM, and GSD of measured specific activities in geotectonical units

Geotectonical units	N	^{40}K [Bqkg^{-1}]		^{226}Ra [Bqkg^{-1}]		^{238}U [Bqkg^{-1}]		^{232}Th [Bqkg^{-1}]	
		Range	GM(GSD)	Range	GM(GSD)	Range	GM(GSD)	Range	GM(GSD)
KZA	8	378-783	639(1.25)	36-101	58(1.36)	43-79	56(1.21)	40-67	52(1.18)
PE	36	193-959	622(1.34)	36-123	55(1.35)	22-111	52(1.41)	35-145	56(1.40)
SMM	25	348-1390	627(1.41)	23-99	40(1.43)	24-108	43(1.46)	23-83	40(1.47)
VZ	90	80-1089	463(1.51)	9-87	29(1.47)	9-82	30(1.48)	7-79	30(1.52)
WMM	54	186-974	621(1.37)	18-86	39(1.38)	15-72	41(1.42)	17-69	40(1.36)

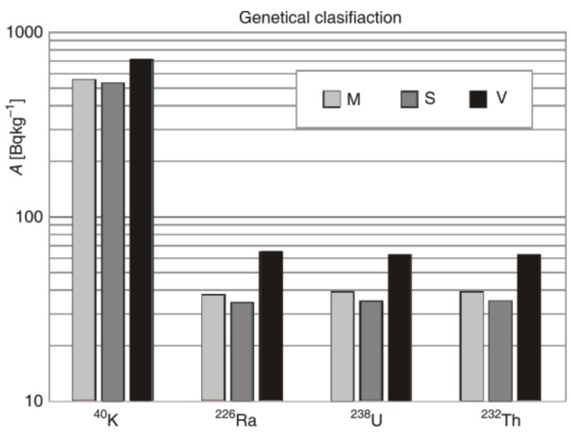

Figure 4. The GM values of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities measured in the soils of sedimentary (S), methamorfic (M), and volcanic origin (V)

ogy. Considering that the level of activities are related to the radionuclides contents in the rock from which soil originated and to the geological composition of each lithologically specific area, we classified our data according to geotectonic units of rock genesis and lithostratigraphy. The corresponding results are given in tabs. 3 and 4, and fig. 4.

Kruskal-Wallis (KW) and Mann-Whitney (MW) tests demonstrated that variations between geological classification for natural radionuclides were significant at a significance level of 95 %, ($p < 0.05$). The results of specific activities per region, given in tab. 3, form two groups for ^{40}K isotope and three groups for ^{226}Ra, ^{238}U, and ^{232}Th. The following significant differences between grouped specific activities were obtained:

^{40}K: VZ < KZA, PE, SMM, WMM

Table 4. Range, GM, and GSD of measured specific activities in lithostratigraphic units

Lithostratigraphic units	N	^{40}K [Bqkg^{-1}]		^{226}Ra [Bqkg^{-1}]		^{238}U [Bqkg^{-1}]		^{232}Th [Bqkg^{-1}]	
		Range	GM(GSD)	Range	GM(GSD)	Range	GM(GSD)	Range	GM(GSD)
Alluvium	63	80-974	549(1.55)	9-99	35(1.59)	9-108	36(1.65)	77-88	36(1.66)
Clay, sand, and gravel	26	186-1071	512(1.44)	20-55	37(1.33)	16-59	36(1.35)	22-68	35(1.33)
Congl., marble and clay	6	278-695	478(1.53)	19-38	28(1.32)	18-61	37(1.49)	17-54	33(1.65)
Congl., sand, marble, and clay	9	295-604	407(1.31)	18-38	24(1.27)	18-42	28(1.33)	9-33	25(1.49)
Diluvium-proluvium	26	400-1089	574(1.25)	18-106	36(1.51)	16-76	37(1.44)	17-64	37(1.40)
Marble and limestone	4	646-735	692(1.05)	36-42	39(1.07)	29-62	39(1.39)	33-53	45(1.23)
Marble and gneiss	5	193-959	519(1.95)	28-90	47(1.62)	22-86	42(1.69)	37-104	59(1.56)
Marble, sand, and clay	3	430-567	493(1.15)	23-26	24(1.07)	21-26	23(1.13)	25-31	28(1.12)
Phyllite	4	509-860	737(1.28)	21-86	42(1.78)	31-64	41(1.37)	24-79	40(1.66)
Philite and shists	8	359-965	600(1.47)	31-52	38(1.23)	42-62	50(1.16)	32-64	46(1.23)
Sandstone, clay, and marble	4	418-596	492(1.17)	17-33	26(1.35)	21-34	28(1.25)	20-32	27(1.23)
Gneiss	9	475-833	603(1.23)	35-73	47(1.29)	36-74	47(1.31)	27-79	41(1.66)
Granit	2	738-899	815(1.15)	58-72	65(1.17)	54-83	67(1.36)	54-78	65(1.66)
Granodiorit	4	508-850	739(1.28)	55-75	66(1.15)	59-64	61(1.04)	41-66	51(1.66)
Ignimbrite	5	626-828	718(1.13)	43-87	61(1.29)	52-82	62(1.19)	51-67	58(1.66)
Limestone	7	297-804	573(1.40)	20-60	33(1.51)	21-56	38(1.40)	23-56	36(1.66)
Marble	6	119-680	348(1.90)	18-47	31(1.37)	15-30	25(1.30)	23-34	30(1.66)
Quartzit	2	580-716	644(1.16)	37-42	39(1.09)	43-47	45(1.06)	37-38	37(1.66)
Rhyolit	2	845-1390	1084(1.42)	46-66	55(1.29)	60-69	64(1.10)	56-82	68(1.66)
Schist	5	372-626	483(1.21)	22-30	26(1.14)	19-52	30(1.49)	23-34	26(1.66)
Tuffite of andesite	8	378-808	605(1.30)	36-123	69(1.47)	43-111	62(1.39)	40-145	68(1.56)
Yellow sandstone	5	272-685	461(1.39)	34-42	36(1.09)	22-43	32(1.30)	22-42	32(1.27)

^{226}Ra: VZ < SMM,WMM < KZA, PE
^{238}U: VZ < SMM,WMM < KZA, PE
^{232}Th: VZ < SMM,WMM < KZA, PE.

The GM for ^{40}K ranged from 463 Bqkg^{-1} in Vardar Zone up to 639 Bqkg^{-1} in Kratovsko Zletavska area. The GM for ^{226}Ra ranged from 29 Bqkg^{-1} in Vardar Zone to 58 Bqkg^{-1} in Kratovsko Zletavska area. The GM for ^{232}Th ranged from 30 Bqkg^{-1} in Vardar Zone to 56 Bqkg^{-1} in Pelagonia.

Figure 4 demonstrates higher radionuclide activities in soil with volcanic (V) origin in comparison to the soils of sedimentary (S), and metamorphic (M) origin. The corresponding GM values for specific activities in soil with metamorphic, sedimentary, and volcanic origin were found to be: 553 Bqkg^{-1}, 529 Bqkg^{-1} and 712 Bqkg^{-1} for ^{40}K; 38 Bqkg^{-1}, 34 Bqkg^{-1}, and 65 Bqkg^{-1} for ^{226}Ra; 40 Bqkg^{-1}, 35 Bqkg^{-1}, and 63 Bqkg^{-1} for ^{238}U; 40 Bqkg^{-1}, 35 Bqkg^{-1}, and 62 Bqkg^{-1} for ^{232}Th, respectively.

The analysis of the results grouped by lithostratigraphic units, tab. 4, revealed the influence on the variation of the activities, but on the other hand, clear grouping was not achieved as in the previous two cases, meaning that some of the values indicated belonging to both groups. Generally, the specific activities can be sorted as follows:

^{40}K: Conglomerates, sandstones, marls and clay < marble and limestone; ignimbrite; phyllite; granodiorites; granites < rhyolites,

^{226}Ra: Marls, sand and clay; conglomerates, sandstones, marls and clay; schist; sandstones, clay and marls; conglomerates, marls and clay; marbles< ignimbrite < tuff of andesite; granites; granodiorites.

^{238}U: Marls, sand and clay; marbles; sandstones, clay and marls; conglomerates, sandstones, marls and clay < phyllite and schist < tuff of andesite; ignimbrite; granites; granodiorites; rhyolites.

^{232}Th: schist < phyllite and schist; granodiorites; marbles and gneisses < tuff of andesite; ignimbrite; granites; rhyolites.

Radionuclides co-occurrence

To draw conclusions about the co-occurrence of the radionuclides among all sampling locations, the correlation between ^{40}K, ^{137}Cs, ^{226}Ra, ^{238}U, and ^{232}Th specific activities were analysed against each other at 95 % significance level. The Spearman coefficients of correlation (ρ), used as a measure of the strength of association are given in tab. 5.

Positive correlations between all natural radionuclides with different levels of association were obtained , tab. 5. The ρ values referring to the correlations between ^{226}Ra, ^{238}U, ^{232}Th were very similar. Considering that ^{238}U and ^{232}Th chains commonly occur together in the environment, these results are expected. However, correlation coefficients with ^{40}K are somewhat lower, indicating the possibility of its different behaviour in soil compared to other natural radionuclides.

For comparison, significant positive correlations with the different level of relations between ^{226}Ra, ^{238}U, and ^{232}Th specific activities were also reported for soils collected in the Central Spanish Pyrenees [35]. In the same study the correlations between ^{40}K and ^{226}Ra, ^{232}Th were stronger than the correlations between ^{226}Ra, ^{238}U, ^{232}Th. On the other hand, in Slovenian soils, moderate positive correlations were observed between ^{232}Th and ^{226}Ra (r = 0.51) and between ^{232}Th and ^{40}K (r = 0.56), and a weak positive correlation between ^{226}Ra and ^{40}K (r = 0.30) [36]. In the soil of Kumaun Himalaya, India, positive correlations were observed between ^{40}K and ^{226}Ra (r = 0.6), ^{40}K and ^{232}Th (r = 0.7), ^{226}Ra and ^{232}Th (0.7) [37].

Generally, different levels of correlation are due to different chemical and physical properties of the elements but, also affected by the differences in their mobility and transfer between environmental compartments. Many other authors have come to this conclusion by examining the specific activities of radionuclides in different soils with different characteristics. For example, the different contents of clay and sand, in soil samples, together with micro-cracks, fractures and mobility characteristic of the radionuclides in a geological medium, also contribute to the difference in the correlation coefficients [37].

Another way of characterizing co-occurrence of different radionuclides on the same location relies on the ratios of specific activities. The distribution of the ratio values in a particular region can give indications of the existence of enrichment or depletion of a particular radioisotope. The basic statistic for the ratios: ^{238}U/^{226}Ra, ^{232}Th/^{226}Ra, ^{40}K/^{226}Ra, and ^{40}K /^{232}Th is presented in tab. 6.

Table 5. Correlation matrix between the specific activities of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th

Variables	^{40}K	^{226}Ra	^{238}U	^{232}Th
^{40}K	1.00	0.63	0.61	0.65
^{226}Ra	0.63	1.00	0.77	0.80
^{238}U	0.61	0.77	1.00	0.81
^{232}Th	0.65	0.80	0.81	1.00

All values significant at significance level: = 0.05

Table 6. Minimum, maximum, GM and GSD of specific activity ratios in soil samples

Ratio	N	Min	Max	GM	GSD
^{238}U/^{226}Ra	213	0.42	2.16	1.02	1.29
^{232}Th/^{226}Ra	213	0.39	1.88	1.02	1.29
^{40}K/^{226}Ra	213	4.25	36.30	14.79	1.44
^{40}K/^{232}Th	213	3.86	42.29	14.55	1.41

The results in tab. 6 show that the ratio of ^{238}U and ^{232}Th to ^{226}Ra is close to unity. The ^{238}U/^{226}Ra is expected to be 1, because both radionuclides are part of the same ^{238}U chain of decay. Deviations from unity may be explained (1) by measurement uncertainty and (2) by true deviation from equilibrium owing to environmental fractionation processes, in particular, due to a different solubility of U and Ra. The mean value of ^{238}U/^{226}Ra in this work is similar to the mean value of 0.95 obtained in a survey in Northern India [38]. On the other hand, it is higher than reported for Central Spanish Pyrenees soils, ^{238}U/^{226}Ra = 0.55 [35].

The same ratio was obtained for ^{232}Th/^{226}Ra, which indicates that in the examined soils, the radionuclides from the two chains are roughly equal in their contribution. Because of this, the ratios of ^{226}Ra and ^{232}Th to ^{40}K were also similar. On the other hand, the intervals of the ratios indicated that, at certain locations, a disturbance of the equilibrium exists. On a global scale, crust abundance of Th is higher than the one of U. The ^{40}K/^{226}Ra obtained in this study ranged from 4.25 to 36.3 with a geometric mean value of 14.8. These results are higher than those for soils collected in India, which varied from 3.4 to 10.15 with a mean value of 6.61 [38].

In order to investigate the relation of the ratios with geology, analysis of variance of the ratios in relation to the geotectonic units, genetic classification and lithostratigraphy were carried out. The analysis showed that only the ratios of ^{226}Ra and ^{232}Th to ^{40}K were different among the geotectonic units. The obtained GM's for ^{40}K/^{226}Ra and ^{40}K/^{232}Th were found to be lower in KZA and PE than in the other geotectonic units. Also, the same was obtained for the genetic classification: in the soils of volcanic origin the ratio was lower in comparison to the soils of sedimentary and methamorphic origin. If lithostratigraphy is considered, ^{40}K/^{226}Ra was significantly classified, but no distinct groups can be identified. The latter means, either, that a soil classification according to the geological parameters is insufficient for understanding co-occurrence process of the investigated radionuclides, or that the number of samples is not sufficient for the analysis. In order to solve this, further investigations of radionuclide variations related to the soil physical and chemical properties, are needed.

CONCLUSIONS

The results of the survey of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th specific activities in surface soils, from 213 sampling sites in the Republic of North Macedona and their relation to geological formations, are presented.

The values of the measured specific activities of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th were found to be in the ranges of the values reported for East and South European countries in the UNSCEAR, 2000 report. The results add to better knowledge of the radiological baseline of North Macedonia, which is the primary purpose of the study, but also contribute to the global geochemical database.

The results were used to map the specific activities for each radionuclide separately, in order to allocate the regions with elevated background values.

Higher levels of ^{40}K, ^{226}Ra, ^{238}U, and ^{232}Th activity concentration were associated mainly with volcanic geology in both, the Pelagonia and Kratovsko Zletovska areas.

Strong positive correlation was evident between ^{238}U and ^{232}Th (ρ = 0.81), ^{238}U and ^{226}Ra (ρ = 0.77), ^{232}Th and ^{226}Ra (ρ = 0.80), good correlations: for the ^{238}U and ^{40}K (ρ = 0.61), ^{232}Th and ^{40}K (ρ = 0.65), ^{40}K and ^{226}Ra (ρ = 0.63).

This study indicates the need of further investigation which should include physical and chemical properties of the soils that may influence radionuclides migration and accumulation in the soil. In order decide about classification power of lithostratigraphy, more samples should be available for a number of units. In certain geological areas with elevated radiometric indicators (geochemical concentration, dose rate), or where geological knowledge suggests so, one may consider undertaking finer sampling in order to detect possible radiation anomalies.

AUTHORS' CONTRIBUTIONS

The manuscript was written by Z. Stojanovska with contribution of all the authors. The figures were prepared by Z. Stojanovska and P. Bossew. The survey was organized by Z. Stojanovska and M. Ristova, soil sampling and gamma spectrometry analysis were carried out by Z. Stojanovska. Explanation of geology was carried out by B. Boev, G. Dimov and I. Boev. In results discussion and review of the manuscript all authors were involved.

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АНАЛИЗА ВАРИЈАЦИЈА СПЕЦИФИЧНЕ АКТИВНОСТИ РАДИОНУКЛИДА У ЗЕМЉИ У ГЕОТЕКТОНСКИМ ЈЕДИНИЦАМА РЕПУБЛИКЕ СЕВЕРНЕ МАКЕДОНИЈЕ

На читавој територији Републике Северне Макдоније извршено је истраживање са циљем утврђивања основних вредности концентрација терестријалних радионуклида. Прикупљено је 213 узорака земљишта са подручја у околини главних насеља и градова, равномерно распоређених по геотектонским јединицама које чине геолошку основу земље. Специфичне активности радионуклида у сувим узорцима земљишта мерене су гама спектрометријом. Добијене су следеће геометријске средње вредности и геометријске стандардне девијације: 550 $Bqkg^{-1}$ (1.47) за ^{40}K, 37 $Bqkg^{-1}$ (1.53) за ^{226}Ra, 38 $Bqkg^{-1}$ за ^{238}U, и 38 $Bqkg^{-1}$ (1.53) за ^{232}Th у сувој земљи. Испитивана је веза између специфичних активности природних радионуклида и геологије. Корелација између радионуклида и геологије, која може служити као додатни геохемијски индикатор, није била убедљива. Утврђене су међусобне корелације између ^{226}Ra, ^{238}U и ^{232}Th активности (Spearman ρ око 0.8), док су оне између њих и ^{40}K мало ниже, $\rho = 0.6$.

Кључне речи: ^{40}K, ^{226}Ra, ^{238}U, ^{232}Th, гама спектрометрија, земља, геологија, литостратиграфска јединица