

DISTRIBUTION OF SOME RADIONUCLIDES IN CORN AND SOIL FROM DIFFERENT LOCATIONS IN THE SURROUNDING OF SKOPJE

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A b s t r a c t: Concentration of natural radionuclides in soil and plants are crucial parameters for defining the level of contamination of the environment. The samples for analysis were collected from the 14 localities of the surrounding of Skopje, of cultivated and uncultivated soil. The specific activity of radionuclides ²³²Th, ²²⁶Ra and ⁴⁰K are determined by gamma spectrometry. The analysis results show that the mean value of ²³²Th specific activity in cultivated soil was 43.74 ± 4.99 Bq·kg⁻¹, while in uncultivated soil it was 35.70 ± 5.65 Bq·kg⁻¹. The ²²⁶Ra specific activity had a mean value of 34.09 ± 6.53 Bq·kg⁻¹ and 28.30 ± 5.54 Bq·kg⁻¹ in cultivated and uncultivated soil, respectively. The specific activity of ⁴⁰K from all localities was higher than the one of ²³²Th and ²²⁶Ra, and in cultivated soil the mean value was 608.12 ± 70.43 Bq·kg⁻¹, while in uncultivated soil it was 506.54 ± 59.96 Bq·kg⁻¹. From the same locations corn samples were also collected for analysis. Concentration of natural radionuclides in corn is lower than referent values. From the results transfer factor (TF) of natural radionuclides from soil to corn was calculated.

Key words: soil; corn; radionuclides; gamma spectrometry; transfer factor

ДИСТРИБУЦИЈА НА РАДИОНУКЛИДИ ВО ПЧЕНКА И ЗА ПОЧВА ОД РАЗЛИЧНИ ЛОКАЛИТЕТИ ОД ОКОЛИНАТА НА СКОПЈЕ

A p s t r a k t: Концентрацијата на природните радионуклиди во почвата и растенијата се основни параметри за дефинирање на степенот на контаминацијата на животната средина, како и на радијациониот ризик за населението. Примероци за анализа се земени од 14 локалитети од околината на Скопје, од обработлива и необработлива почва. Специфичната активност на радионуклидите ²³²Th, ²²⁶Ra и ⁴⁰K е определена со гама-спектрометрија. Средната вредност на специфичната активност на ²³²Th во обработлива почва изнесува 43.74 ± 4.99 Bq·kg⁻¹, а во необработлива почва 35.70 ± 5.65 Bq·kg⁻¹. Специфичната активност на ²²⁶Ra има средна вредност од 34.09 ± 6.53 Bq·kg⁻¹ и 28.30 ± 5.54 Bq·kg⁻¹ за обработлива и необработлива почва соодветно. Специфичната активност на ⁴⁰K на сите локалитети е повисока во однос на ²³²Th и ²²⁶Ra, и за обработливата почва средната вредност изнесува 608.12 ± 70.43 Bq·kg⁻¹, а за необработливата почва изнесува 506.54 ± 59.96 Bq·kg⁻¹. Од истите места се земени и примероци од пченка за анализа. Концентрацијата на радионуклидите во пченката е пониска во однос на референтните вредности. Од добиените резултати е пресметан и факторот на трансфер (ТФ) на природните радионуклиди од почвата во пченката.

Клучни зборови: почва; пченка; радионуклиди; гама-спектрометрија; радијационен ризик

1. INTRODUCTION

Dominantly radionuclides are accumulated in the soil surface. Migration of the radionuclides in the soil depends on various factors, dominantly by the soil properties, concentration of stable isotopes,

as well as the intensity of the agricultural activity [1–3].

The aim of this research is to define concentration of natural radionuclides in soil [4, 5]. The specific activity of ²²⁶Ra, ²³²Th and ⁴⁰K is determined

on samples collected from cultivated and uncultivated soil [6–9].

Radionuclides in soil are available for the plants [13–24]. Also the corn extracting the radionuclides from soil. Corn is used as food in the human diet and also as animal feed. Consumption of contaminated corn may cause serious health disorders. Therefore the concentration of natural radionuclides in corn is determined. Also the transfer factors of radionuclides from soil to corn are defined [25, 26].

2. MATERIALS AND METHODS

The research area is the surrounding of Skopje. The total area is 2100 km² with an altitude between 173 and 340 m. The average annual air temperature was 13.7 °C, with average annual rainfalls of 412.7 mm [27].

The soil samples for analysis were collected in 2014 from 14 localities in the surrounding of Skopje (Figure 1). The samples were collected from cultivated and uncultivated soil, by appropriate soil sampling tube at three various depths: 0 – 5 cm, 5 – 10 cm and 10 – 15 cm. The soil samples were collected in duplicates, six soil samples from each locality. During the sampling, a special care was taken for the micro-location flatness, in order to exclude the possibility for horizontal radionuclide translocation.

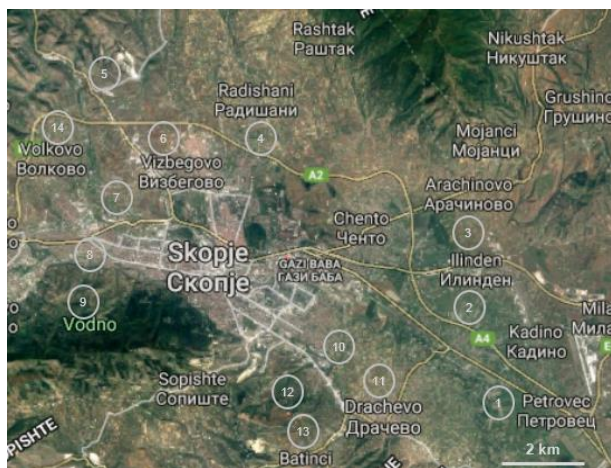


Fig. 1. Sampling localities of soil from the surrounding of City of Skopje: 1 – Petrovec, 2 – Ilinden, 3 – Aračinovo, 4 – Radišani, 5 – Chucher Sandevo, 6 – Vizbegovo, 7 – Bardovci, 8 – Saraj, 9 – Nerezi, 10 – Lisiče, 11 – Dračevo, 12 – Pintija, 13 – Batinci, 14 – Volkovo

After removing the stones and organic impurities, the samples were dried in an oven at 110 °C for

24 hours, to remove the moisture. Then, the samples were crushed and passed through 150 μm sieve. A mass from 480 to 490 g of each homogenized sample was weighed and packed in polyethylene Marinelli containers. After tightening, the containers were sealed with adhesive tape and left for 21 days before measuring with gamma spectrometry. This was in order to ensure that the daughter products of ²²⁶Ra and ²²⁸Th have achieved equilibrium with the respective parent radionuclides.

The gamma spectrometry was used for determination of radioactivity of the soil samples [28, 29]. The spectrometer consisted of an HPGe detector, model 3020 (Canberra Packard, Meriden, CT, USA), with active volume of 180 cm³, relative efficiency of 30 %, operating voltage 3000 V, and resolution of 2 keV at 1332.5 keV. Data acquisition and analysis were performed with 8192 channel digital analyzer; duration of acquisition interval for each sample was 65 000 s. The activity of ²²⁶Ra was determined from the gamma lines associated with low half-live time daughters of ²¹⁴Bi (609.31, 1120.29 and 1794.49 keV) and ²¹⁴Pb (351.93 keV). The ²³²Th activity was determined by 338.4, 911.2 and 969.1 keV gamma lines from ²²⁷Ac and its decay products. The gamma line at 1460.8 keV was used to determine the activity of ⁴⁰K.

Efficiency calibration was performed with mixed calibration standard sources MBSS2, supplied from the Czech Metrological Institute, Inspectorate for Ionizing Radiation. In order to determine the background distribution in the detector environment, empty sealed Marinelli beaker with the same geometry was measured at equal counts as the soil samples. The analysis procedure included the subtraction of the background spectrum.

The specific activity (*A*) is determined according the equation [12]:

$$A = \frac{N - N_0}{\varepsilon \cdot \gamma \cdot t \cdot m} \quad (\text{Bq} \cdot \text{kg}^{-1}),$$

where *N* is clean surface of peak accumulated from a specific radionuclide in analysis of a specific sample (number of readings), *N*₀ is clean surface of peak accumulated from the spot of a specific radionuclide without an analysis of sample (number of readings), *t* is live time of accumulation of the sample spectrum (s), *t*₀ is live time of accumulation of the phone spectrum (s), *ε* is detector efficiency for a given energy (for a specific peak), *γ* is intensity of gamma transition in radioactive decay for a respective radionuclide (%), and *m* is sample mass (kg).

Corn samples were analyzed from the same locations, 3 samples from each location, which is in accordance with the International Atomic Energy Agency recommendations.

Transfer of radionuclides from soil to plant is determined by transfer factor – TF [25, 26]. This factor include radionuclide concentration per gram of plant (dry or wet plant weight) ($\text{Bq}\cdot\text{kg}^{-1}$) at harvest divided by radionuclide concentration per gram of soil ($\text{Bq}\cdot\text{kg}^{-1}$). TF is dependent on the factors such as: radionuclide type, product type, soil type, EC, pH and bicarbonate content of the soil. Using TF we can find if plants receive contamination from soil or root.

$$\text{TF} = \frac{\text{radionuclide concentration in plant (dry weight)}}{\text{radionuclide concentration in soil (dry weight)}}$$

3. RESULTS AND DISCUSSION

In Tables 1, 2 and 3 the specific activities of radionuclides of ^{226}Ra , ^{232}Th and ^{40}K are presented for uncultivated and cultivated soil samples. The obtained results are from six measurements at three various soil depths (in duplicate). The values are associated with the respective standard deviations (SD).

Table 1

Comparison results from measured specific activities (A) of ^{226}Ra

Sampling sites	A±SD ($\text{Bq}\cdot\text{kg}^{-1}$)		Increased A %
	Uncultivated soil	Cultivated soil	
Petrovec	28.22±1.51	34.36±4.19	21.76
Ilinden	26.01±1.53	32.41±3.25	24.61
Aračinovo	33.79±1.82	41.21±1.75	21.96
Radišani	27.32±4.20	31.39±1.18	14.90
Čučer-Sandev	23.86±1.31	24.06±0.56	0.83
Vizbegovo	36.95±2.55	40.78±1.02	10.37
Bardovci	39.43±4.16	41.91±5.61	6.30
Saraj	32.32±4.52	36.03±0.54	11.48
Nerezi	27.62±1.52	36.33±1.32	31.54
Lisiče	20.38±1.40	26.27±1.37	28.90
Dračevo	22.64±2.05	25.73±1.64	13.65
Pintija	30.69±1.42	40.87±1.23	33.17
Batinci	27.12±1.32	36.01±4.21	32.78
Volkovo	23.70±1.51	29.30±5.14	23.63

Table 2

Comparison results from measured specific activities (A) of ^{232}Th

Sampling sites	A±SD ($\text{Bq}\cdot\text{kg}^{-1}$)		Increased A %
	Uncultivated soil	Cultivated soil	
Petrovec	28.27±2.44	42.42±4.23	50.05
Ilinden	32.71±1.22	42.45±1.18	29.78
Aračinovo	45.67±4.44	48.08±1.16	5.27
Radišani	39.61±1.21	51.47±2.67	29.94
Čučer-Sandev	33.90±3.40	38.54±2.50	13.69
Vizbegovo	38.12±2.52	49.57±1.67	30.04
Bardovci	42.92±4.50	49.82±2.21	16.08
Saraj	40.32±2.25	40.96±1.91	1.59
Nerezi	38.40±2.02	52.19±1.63	35.91
Lisiče	34.73±2.27	40.80±2.05	17.48
Dračevo	28.57±4.35	39.87±1.47	39.55
Pintija	33.57±1.91	45.96±3.30	36.91
Batinci	35.97±2.30	42.35±1.21	17.74
Volkovo	27.07±2.32	41.90±3.63	54.78

Table 3

Comparison results from measured specific activities (A) of ^{40}K

Sampling sites	A±SD ($\text{Bq}\cdot\text{kg}^{-1}$)		Increased A %
	Uncultivated soil	Cultivated soil	
Petrovec	439.94±20.12	587.59±11.20	33.56
Ilinden	489.63±20.84	573.65±16.41	17.16
Aračinovo	540.24±19.69	676.71±18.18	25.26
Radišani	507.19±20.30	622.56±9.09	22.75
Čučer-Sandev	450.80±21.46	501.86±55.10	11.33
Vizbegovo	605.29±44.91	704.54±16.95	16.40
Bardovci	625.81±48.21	706.86±13.63	12.95
Saraj	476.45±22.09	628.70±19.16	31.96
Nerezi	522.71±20.12	623.47±16.19	19.28
Lisiče	437.26±45.55	526.76±38.58	20.47
Dračevo	431.45±21.25	549.21±24.17	27.29
Pintija	522.60±21.74	655.97±25.56	25.52
Batinci	488.56±20.09	629.50±31.58	28.85
Volkovo	476.07±20.09	526.27±72.05	10.54

The variations in the specific activity values of radionuclides of ^{226}Ra , ^{232}Th and ^{40}K are due to the various physical and geochemical soil properties. As expected, and in accordance with the worldwide data [30, 31], the specific activities of ^{232}Th are

higher than the ones of ^{226}Ra , while the values of the specific activities of ^{40}K are one magnitude higher.

The statistical parameters are calculated and presented in Table 4.

Table 4

Statistical data analysis of ^{226}Ra , ^{232}Th and ^{40}K

	^{226}Ra		^{232}Th		^{40}K	
	Uncultivated soil	Cultivated soil	Uncultivated soil	Cultivated soil	Uncultivated soil	Cultivated soil
Mean value (Bq·kg ⁻¹)	28.58	34.05	35.70	44.74	501.00	608.12
Median (Bq·kg ⁻¹)	27.47	35.19	35.35	42.44	489.10	623.02
Minimum (Bq·kg ⁻¹)	20.38	24.06	27.07	38.54	431.45	501.86
Maximum (Bq·kg ⁻¹)	39.43	41.91	45.67	52.19	625.81	706.86
SD (Bq·kg ⁻¹)	5.49	6.53	5.61	4.99	61.80	70.43
Range (max-min)	19.05	17.85	18.60	13.65	194.36	205.00
Skewness	0.58	-0.27	0.05	0.42	0.94	-0.07
Kurtosis	-0.28	-1.17	-0.65	-1.42	0.38	-1.05

For cultivated soils, the mean value of the specific activities of ^{226}Ra is lower than the worldwide average value (35 Bq·kg⁻¹). The calculated mean value of ^{232}Th is higher than the world average value (30 Bq·kg⁻¹). Also the calculated mean value of the specific activities of ^{40}K is higher than the worldwide average value (400 Bq·kg⁻¹) [30, 31]. According to presented results for uncultivated soil, the mean values of the specific activities of ^{226}Ra is lower than the world average value. The calculated mean values of the specific activities of ^{232}Th and ^{40}K are higher than the world average values [30, 31].

There are significant variations between the two data series of activities measured for uncultivated and cultivated soil samples. The specific activity values of radionuclides of ^{226}Ra , ^{232}Th and ^{40}K are higher in cultivated soil as a consequence of fertilization by agricultural activity [10–12]. The main source for the increased levels of natural radionuclides are phosphate and potassium fertilizers. According to literature data, the levels of ^{226}Ra , ^{232}Th and ^{40}K in fertilizers may vary up to 1852 Bq·kg⁻¹, 65 Bq·kg⁻¹ and 4860 Bq·kg⁻¹. Also the statistical values are calculated (Table 5).

Plants don't have equal affinity for extraction of present radionuclides in soils. The specific activ-

ities (A) values of ^{226}Ra , ^{232}Th and ^{40}K in corn samples are presented in Table 6. The specific activity value of radionuclide of ^{40}K is the highest, because K is a major nutrient for plants.

The transfer factor soil to corn is also calculated (Table 7). The transfer factor values of ^{226}Ra are higher than the ^{232}Th , as a consequence of difference in solubility. Because K is a major nutrient, ^{40}K has the highest values for soil-to-corn TF in comparison with radionuclides of ^{226}Ra and ^{232}Th [25, 26].

Table 5

Calculated statistical increasing

	^{226}Ra	^{232}Th	^{40}K
Mean value (Bq·kg ⁻¹)	19.71	27.06	21.67
Median (Bq·kg ⁻¹)	21.86	29.86	21.61
Minimum (Bq·kg ⁻¹)	0.83	1.59	10.54
Maximum (Bq·kg ⁻¹)	33.17	54.78	33.56
SD (Bq·kg ⁻¹)	10.29	15.82	7.45
Range (max-min)	32.34	53.19	23.02
Skewness	-0.29	0.11	0.01
Kurtosis	-0.97	-0.68	-1.10

Table 6

Specific activities (A) of ^{226}Ra , ^{232}Th and ^{40}K for corn samples

Sampling sites	A±SD (Bq·kg ⁻¹)		
	^{226}Ra	^{232}Th	^{40}K
Petrovec	0.04±0.10	0.06±0.02	108.15±2.00
Ilinden	1.30±0.40		99.00±2.00
Aračinovo	0.16±0.04	0.02±0.12	69.30±1.00
Radišani	0.73±0.42	0.08±0.03	85.35±1.50
Čučer-Sandevo	0.17±0.11		99.86±1.54
Vizbegovo	0.40±0.05	0.17±0.20	115.54±2.40
Bardovci	0.08±0.03		115.26±2.00
Saraj	0.29±0.20	0.15±0.03	92.00±1.70
Nerezi	0.38±0.20	0.10±0.40	89.57±1.50
Lisiče	0.61±0.21	1.02±0.45	117.21±2.50
Dračevo	0.48±0.20	0.30±0.25	78.02±2.00
Pintija	0.61±0.25	0.11±0.20	195.97±2.00
Batinci	0.33±0.15	0.17±0.05	107.50±2.35
Volkovo	0.62±0.08	0.94±0.04	112.48±2.50

Table 7

Transfer factor (TF) soil to corn

Sampling sites	^{226}Ra	^{232}Th	^{40}K
Petrovec	0.011	0.001	0.184
Ilinden	0.040	<MDA	0.172
Aračinovo	0.003	0.001	0.102
Radišani	0.023	0.001	0.137
Čučer-Sandevo	0.007	<MDA	0.198
Vizbegovo	0.009	0.003	0.163
Bardovci	0.001	<MDA	0.163
Saraj	0.008	0.003	0.146
Nerezi	0.010	0.001	0.143
Lisiče	0.023	0.025	0.222
Dračevo	0.018	0.007	0.142
Pintija	0.014	0.002	0.298
Batinci	0.009	0.004	0.170
Volkovo	0.021	0.022	0.213

4. CONCLUSIONS

The gamma spectrometry was used to determine the concentration of natural radionuclides ^{226}Ra ,

^{232}Th and ^{40}K in soil from 14 localities of the surrounding of Skopje. The results show that the mean value of specific activity of ^{232}Th is 44.74 Bq·kg⁻¹ in cultivated soil, while in uncultivated soil is 35.70 Bq·kg⁻¹. The ^{226}Ra specific activity has a mean value of 34.05 Bq·kg⁻¹ and 28.58 Bq·kg⁻¹ in cultivated and uncultivated soil, respectively. The specific activity of ^{40}K from all localities was higher than the ones of ^{232}Th and ^{226}Ra , and in cultivated soil the mean value is 608.12 Bq·kg⁻¹, while in uncultivated soil it is 501.00 Bq·kg⁻¹.

The obtained data indicate that the specific activity of natural radionuclides in uncultivated soil is significantly lower than in cultivated soil. This is a consequence of application of fertilizers on agricultural soil. According to measured values, it can be concluded that the level of natural radioactivity in soil does not pose serious radiation risk for the human population. However, continuous and systematic monitoring is required in order to assess the level of natural radioactivity in soil.

Since corn is used as a basic raw material in the production of feed, the content of natural radionuclides in corn is also determined, and the transfer factor soil-to-corn is defined. The results indicated that radionuclide concentrations in corn are lower, and do not pose serious radiation risk for the population.

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