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Časopis "Poljoprivreda i šumarstvo"

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Ministry of Agriculture and Rural Development of Montenegro

September 2020

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Aleksandra ANGJELESKA, Ljupčo ANGELOVSKI, Risto UZUNOV, Elizabeta DIMITRIESKA STOJKOVIČ, Biljana STOJANOVSKA DIMZOSKA, Zehra HAJRULAI MUSLIU, Ljubomir ARSOV [1]

DETERMINATION OF THE VERTICAL DISTRIBUTION OF ^{226}Ra, ^{232}Th, ^{40}K AND ^{137}Cs IN SAMPLES OF CULTIVATED SOIL TAKEN IN THE VICINITY OF CERTAIN CITIES IN REPUBLIC OF MACEDONIA

SUMMARY

The purpose of this research was focused on determination of the concentrations of activity of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the soil samples collected from cultivated soil close to several larger cities in the Republic of Macedonia (Tetovo, Veles, Bitola) by using gamma spectrometry.

Data show that the average value of activity of ^{226}Ra and ^{232}Th are within the range of (25.58-36.48) and (36.55-50.45) Bq $kg^{-}1$, respectively. While, the concentrations of activity of ^{40}K in comparison to those obtained for ^{232}Th and ^{226}Ra are higher in all analyzed soils samples, showing values within the range of (611.59-781.23) Bq $kg^{-}1$, the activities of ^{137}Cs are generally lower and are within the range of (9.33-18.90) Bq $kg^{-}1$, with the highest activity at a depth of 5 cm, which is well in accordance with the data from the literature.

However, ^{40}K is the only radionuclide which was determined with a significant quantity in the soil samples, and for which one can tell that in all locations at a depth to 5 cm it shows highest level of activity. It is considered that all other determined radionuclides which occur naturally, have nominal concentrations.

On the basis of the obtained results so far, we can conclude that the soil samples which have been collected in the environment of the observed cities, do not indicate increased radioactivity, which could impose a threat for the food production that might have negative influence on human's health. The concentrations of these radionuclides are comparable to the available data from the other countries.

Keywords: radioactivity; soil; radionuclides; gamma spectrometry; specific activity

INTRODUCTION

People have always been exposed to natural radiations which arise from the interior and the exterior of the soil. Radionuclides come to the surface of the soil in a form of solid particles, or with rains, in dissolved or undissolved state.

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The ones that come in a form of solid particles, are being mechanically retained on the surface, while the ones which have been dissolved with the process of filtration, penetrate the soil and most of them are connecting to its surface layer. 137Cs comes in the soil in a form which is dissolvable in water.

Numerous researches have proved that it is mostly retained on the surface of the bottom, and its speed of penetration through the soil would depend on the type of soil and the quantity of atmospheric precipitation [1].

On the basis of numerous researches, a conclusion can be made that the migration of radionuclides in soil depends on most characteristics of the soil such as physical-chemical characteristics (pH, mineral composition, content of organic matter), structure (porosity, mechanical composition), water regime (content of water, level of underground waters), agro-technical measures (fertilization, tillage) etc. [2].

Natural radioactivity is widely spread in the soil environment and it exists in different geological formations in soil, rocks, plants, water and air [3, 4, 5]. Natural radioactivity in the soil samples comes from series of U and Th and natural K. Artificial radionuclides can also be present, such as 137Cs, which is a result of the accident in Chernobyl, and also of the low sedimentation from different atmospheric trials of nuclear weapons in certain areas of the planet.

Monitoring of radioactivity in soil provides information about its nature, hence it is important for the evaluation of the radiation dose to which the general population is exposed. It turned out that the understanding of the behaviour of natural radionuclides in the environment is very important, since such information can be used as the related parameter values about radiological assessments [6]. For this reason the assessment of the dose of gamma radiation from natural resources has great importance since natural radiation is the main contributor for the external dose in world population [7].

Lately, radioactive matters due to their specific characteristics have been subject of numerous researches, and they are one of the main problems for soil protection. Most attention has been paid to artificial radionuclides, and the natural ones have been less important, in terms of research, as well as in terms of protection [8-9]. However, in order to be able to monitor the artificial radioactivity in soil, it is also necessary to know the natural side of the same.

Many scientists in the world have been working on this field i.e. on research of radioactivity in soils by using gamma spectrometry system in order to determine ^{238}U ^{235}U ^{40}K ^{137}Cs [10-11].

The purpose of this research was focused on determination of concentrations of activity of ^{226}Ra, ^{232}Th,^{40}K and ^{137}Cs in the soil samples collected from three larger cities in Republic of Macedonia, by means of the gamma spectrometry method

MATERIAL AND METHODS

In order to measure the natural radioactivity in soil, soil samples have been taken from 3 larger cities in Republic of Macedonia. Each sample has been taken by means of a special dosing vessel with limiters, which allows sampling of samples at a depth of 0-5 cm, 5-10 cm and 10-15 cm, thereby enabling sampling above these soil layers. Soil sampling has been performed by taking 3 samples from every location, for the given depths according to the recommendations by IAEA [12]. The sampling was performed in May and June 2012.

It was insured to have the micro-location as a flat terrain, thereby excluding the consequences for eventual horizontal translocation of radionuclides. Collected samples were carefully cleaned of small stones, and then they were dried in an electrical oven at a temperature of 110°C to a period of 48 hours depending on the soil depth, until the sample gets constant weight. Upon drying, the samples were crushed, set on a foundation and grinded to a previously determined size of particles according to the analytical requirements and then they passed through a sieve. The homogenized soil samples were packed in plastic containers which had the same geometry as the one of the reference materials.

Upon ensuring time balance between the successors of ^{238}U and ^{232}Th series (21 days), these sealed samples were prepared for analysis.

The spectral analysis of radionuclides of these samples has been conducted by applying spectrometer for γ-rays with high purity germanium detector (HPGe) with 30% relative efficiency and energetic resolution (FWHM) of 1.8 keV for 1.33 MeV reference transition of ^{60}Co [13].

The detector has been protected with lead with a thickness of 9 cm with an internal line of 0,5 cm thin copper plate covered with 1 mm aluminium in order to absorb the x-rays from the lead and the copper. The internal size of the shell gap was 30 x 30 x 30 cm. The detector was given high voltage through a preamplifier which was afterwards connected with an amplifier with a computer-based channel analyser through ADC (analogous to digital convertor). The software which was used for obtaining data is Canberra software package Genie-2000, including research of maximal value and modules for identification of nuclides. The system was regularly calibrated for energy and efficiency. The energetic calibration was performed by obtaining a spectrum of approved calibration sources of known energies such as ^{60}CO, for E_{γ}=1332.5 and 1173 keV, and ^{137}Cs, for E_{γ}=661.6 keV. The gamma rays which were the point of interest were within the range from 50 to 3000 keV. The prepared Marinelli glasses (samples) have been placed on a final detector at a distance of approximately 10 mm. Each sample was counted for a period of 65000s in order to get good statistics and the constant time was less than 10%. The measurements with an empty Marinelli glass, during identical conditions, have been also conducted in order to determine the background. Then they have been deducted from the measured spectrums of each sample in order to get the net activities of radionuclides.

The specific activity $A_{E,i}$(Bq/kg) is determined [14],

$$A_{E,i}= N_{E,i}/ \varepsilon_E x t x I_a x m$$

where:

$N_{E,i}$-surface at the top of total energy absorption E,

ε_E-efficiency of the energy detector E,

t-time of measurement,

I_a-absolute intensity of gamma decay of energy E and

m – sample mass (kg).

For the purpose of determination of activity of ^{226}Ra, gamma lines of ^{214}Bi at 609,3 keV and ^{214}Pb at 351,9 keV, have been used. For the purpose of determination of the activity of ^{232}Th, gamma lines of ^{228}Ac have been used at 911,2 and 969,1 keV and the line of ^{214}Pb at 238,6 keV. The activity of ^{40}K has been determined on the basis of the line 1460.8 keV. The activity of ^{137}Cs is on the basis of the line 661.66 keV [15].

RESULTS AND DISCUSSION

Obtained specific activities of the observed radio-nuclides in soil samples taken from locations of cultivated land in the vicinity of several cities in the Republic of Macedonia are presented in the Table 1. The table shows the mean values of the specific activities of the given elements with the appropriate standard deviation.

Data show that the average value of activity of ^{232}Th is within the range from 36.55 Bq kg^{-}1 to 50.45 Bq kg^{-}1 and it is higher than the one of ^{226}Ra, which may be due to a longer half-life of ^{232}Th in relation to ^{226}Ra. The activity of ^{226}Ra is within the range from 25.58 Bq kg^{-}1 to 36.60 Bq kg^{-}1.

The activity of ^{40}K is within the range of 611.59 Bq kg^{-}1 to 781.23 Bq kg^{-}1. The activity of ^{137}Cs is within the range of 9.33 Bq kg^{-}1 to 18.90 Bq kg^{-}1, however the highest values have been measured at a depth to 5 cm.

In the current study, it has been noted that the specific activity of these radionuclides in the soil is not uniform; however it is different for different soils depending on the geological or the typographical character of the area. Additionally, it also depends on the type of the past agricultural activities and different minerals that are present in soil.

One can notice that the average concentrations of the same isotope differ for different soil depths, but common relation between the activity and the depth for ^{226}Ra and ^{232}Th has not been found. The measured values of radioactivity show that it is accidentally distributed in different soil depths which are being examined.

The concentration of activity of ^{40}K in the soil in all locations has higher value than the one of ^{232}Th and ^{226}Ra for all soils which is in accordance with the data from the literature [16]. However, ^{40}K is the only radionuclide determined with an important quantity in the soil samples for which one can say that it shows the highest level of activity in all locations at a depth of 5cm. It is considered that all other determined radio-nuclides which occur naturally have nominal concentrations.

Table 1. Mean values of specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs with the appropriate standard deviation in samples of cultivated soil

Cultivated soil	Specific activities (Bq/kg)				
	Depth (cm)	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Tetovo	0-5	36.25±2.54	51.84±3.63	798.0±55	9.38±0.64
	5-10	37.08±2.60	50.17±3.51	774.91±54	9.15±0.64
	10-15	36.80±2.58	50.44±3.53	794.28±55	9.26±0.65
	0-5	36.40±2.52	51.26±3.50	782.00±54	9.70±0.65
	5-10	36.28±2.60	49.77±3.51	768.20±55	9.18±0.65
	10-15	35.92±2.60	49.70±3.51	759.33±55	9.10±0.63
	0-5	37.05±2.50	50.94±3.50	788.0±55	9.88±0.65
	5-10	36.28±2.50	50.07±3.51	794.11±55	9.26±0.65
	10-15	36.26±2.51	49.84±3.50	772.28±55	9.02±0.65
Veles	0-5	36.68±2.57	47.16±3.30	708.75±49.61	17.77±1.24
	5-10	35.96±2.52	46.37±3.24	695.28±48.67	19.33±1.35
	10-15	37.09±2.60	47.39±3.32	711.76±49.82	19.34±1.35
	0-5	36.02±2.55	46.88±3.33	698.00±49.65	16.81±1.25
	5-10	35.48±2.55	46.27±3.31	672.33±49.65	18.86±1.36
	10-15	36.42±2.51	47.28±3.33	692.00±48.66	18.09±1.25
	0-5	37.08±2.51	46.12±3.31	712.15±49.65	18.27±1.24
	5-10	37.06±2.51	46.52±3.30	703.08±49.65	18.33±1.25
	10-15	37.58±2.51	47.20±3.31	689.00±49.66	17.51±1.25
Bitola	0-5	25.18±1.69	35.96±2.51	619±42.85	19.60±1.37
	5-10	25.06±1.68	38.44±2.68	623.13±43.84	19.89±1.35
	10-15	25.42±1.69	37.39±2.50	593.00±43.00	17.22±1.37
	0-5	26.20±1.68	35.22±2.50	654.00±43.80	18.77±1.34
	5-10	25.91±1.69	37.03±2.55	643.13±42.85	18.65±1.37
	10-15	25.00±1.69	36.02±2.50	609.00±42.85	18.02±1.35
	0-5	26.50±1.65	36.12±2.51	627.00±43.85	19.77±1.34
	5-10	25.75±1.65	36.08±2.50	599.85±43.85	19.65±1.35
	10-15	25.16±1.64	36.72±2.50	536.20±43.85	18.53±1.35

Table 2. Descriptive statistics of the mean values of the specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in Tetovo

Cultivated soil/Tetovo	Specific activity			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Mean value	36.48	50.45	781.23	9.33
Median	36.28	50.17	782.00	9.26
St. deviation	0.40	0.75	13.39	0.29
Minimum	35.92	49.70	759.33	9.02
Maximum	37.08	51.84	798.00	9.88
Rang	1.16	2.14	38.67	0.86
Kurtosis	-0.93	-0.29	-1.19	0.49
Skewness	0.54	0.90	-0.29	1.19

Table 3. Descriptive statistics of the mean values of the specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in Veles

Cultivated soil/Veles	Specific activity			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Mean value	36.60	46.80	698.04	18.26
Median	36.68	46.88	698.00	18.27
St. deviation	0.68	0.48	12.82	0.84
Minimum	35.48	46.12	672.33	16.81
Maximum	37.58	47.39	712.15	19.34
Rang	2.10	1.27	39.82	2.53
Kurtosis	-0.87	-1.91	0.73	-0.44
Skewness	-0.29	-0.19	-0.86	-0.23

Table 4. Descriptive statistics of the mean values of the specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in Bitola

Cultivated soil/Bitola	Specific activity			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Mean value	25.58	36.55	611.59	18.90
Median	25.42	36.12	619.00	18.77
St. deviation	0.54	0.96	34.26	0.91
Minimum	25.00	35.22	536.20	17.22
Maximum	26.50	38.44	654.00	19.89
Rang	1.50	3.22	117.80	2.67
Kurtosis	-1.02	0.73	2.57	-0.37
Skewness	0.62	0.82	-1.29	-0.66

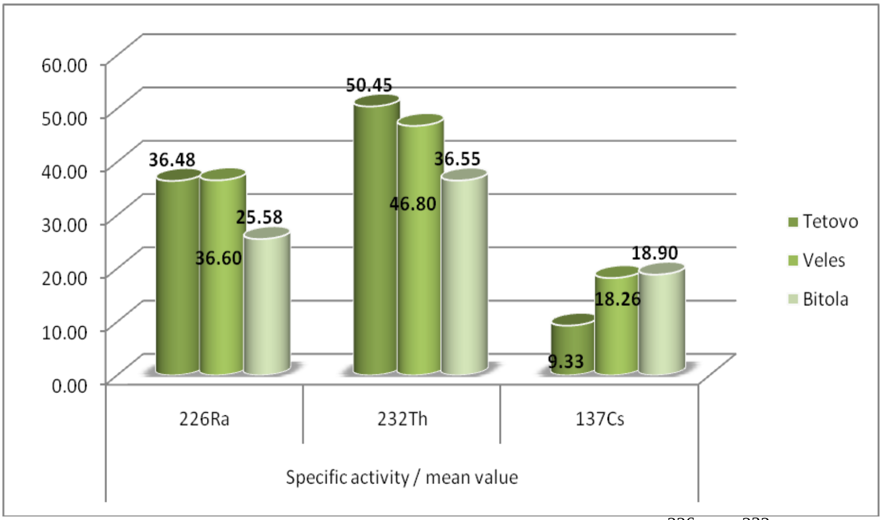

Figure 1. Mean values of the specific activities of ^{226}Ra, ^{232}Th and ^{137}Cs in the three cities in Macedonia

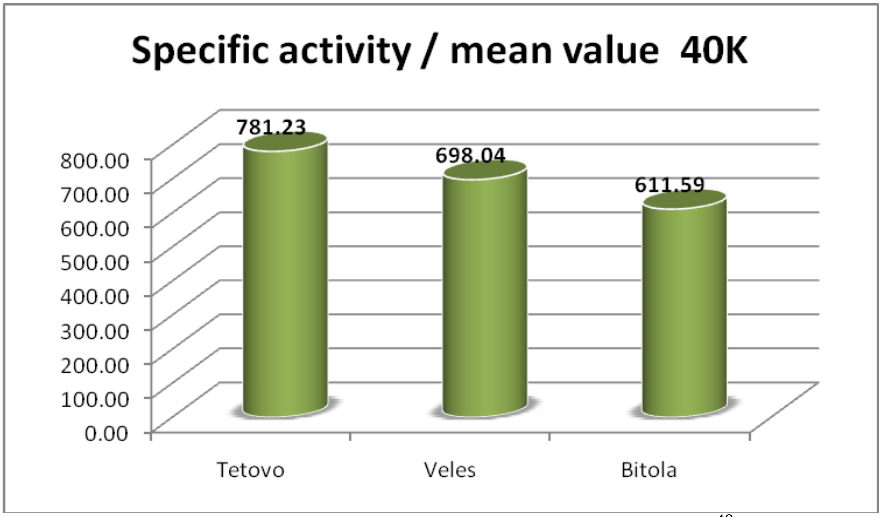

Figure 1. Mean values of the specific activities of ^{40}K
in the surrounding of Skopje and three cities in Macedonia

Different factors may be responsible for the unequal distribution of ^{137}Cs. This variation of data is not very significant taking into consideration the great geographic variations which may be a result of the difference in the characteristics of soil, the ecological and meteorological factors, especially the rain in a period of sedimentation, which is very important because it facilitates sedimentation. However, from the very research one can perceive that in all locations, the highest level of ^{137}Cs is at a depth of 5cm which is in accordance with the data from the literature [17].

All the results from this study have been completely compared to the international values and the ones from other studies in other countries through the entire world. It was determined that the levels of natural radioactivity in Macedonia are not within the range of high risk according to the international standards i.e. the soil samples which have been analysed in the indicated cities do not indicate increased radioactivity which could impose a threat for the food production or which might have negative influence on human’s health.

The results for the natural radioactivity have been compared to the results for different countries from the world. The mean value of specific activity of ^{226}Ra from all locations is 32.88 Bq kg^{-}, which is less than the average value in the world, i.e. 35 Bq kg^{-1}. This level is similar to the one in Romania (32 Bq kg^{-}1), and lower than Croatia (43 Bq kg^{-}1) and Bulgaria (45 Bq kg^{-}1) [7].

The mean value of specific activity of ^{232}Th in all areas of investigation is 44.60 Bq kg^{-1} and it is near the average value in the world, i.e. 45 Bq kg^{-1}. This level of activity is less than in Brazil [18], India, Malaysia, as well as the other areas of Pakistan, i.e. the East Salt Range [19].

This level is higher than in Hungary (28 Bq kg^{-1}) and Bulgaria (30Bq kg^{-1}).

The average level of specific activity of ^{40}K in the soil is 696.95 Bq kg^{-1} which is higher than the average value in the world, i.e. 400 Bq kg^{-1} [20].

This level is higher than in Algeria, Armenia, Bangladesh, China, Cyprus, Denmark, Egypt, Greece, Hungary, India, Iran, Malaysia, Norway, Poland, Sweden, Syria, USA [20] and Brazil [18].

For countries such as Hungary (370 Bq $kg^{-}1$), Bulgaria (400 Bq kg^{-}), Croatia (423 Bq kg^{-}), and Romania (490 Bq $kg^{-}1$), one can say that they have lower level of specific activity of ^{40}K in soil than the mean value measured in the three different cities in Macedonia.

CONCLUSIONS

We can conclude that the soil samples that have been analyzed in the indicated cities do not show increased radioactivity, which would threaten the food production or would exert negative influence on human health. The data obtained with this work can certainly be used as reference values for current estimation of the equivalent dose due to natural radioactivity. However, on the basis of all measured and estimated data, radiation risk does exist, however what is necessary is continuous and systematic examination in order to assess any changes of the level of natural and artificial radioactivity.

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UTVRĐIVANJE VERTIKALNE DISTIBUCIJE ^{226}Ra, ^{232}Th, ^{40}K I ^{137}Cs KOD PRIMJERA KULTIVISANOG ZEMLJIŠTA UZETOG U BLIZINI ODREĐENIH GRADOVA REPUBLIKE MAKEDONIJE

Cilj ovog istraživanja je usmjeren na utvrđivanje koncentracije aktivnosti ^{226}Ra, ^{232}Th, ^{40}K i ^{137}Cs kod uzoraka zemljišta sakupljenog blizu nekoliko većih gradova u Republici Makedoniji (Tetovo, Veles, Bitola), koristeći gama spektometriju.

Podaci pokazuju da je prosječna vrijednost aktivnosti ^{226}Ra i ^{232}Th bila u rasponu od (25,58-36,48) odnosno (36,55-50,45) Bq kg^{-}1. Dok su koncentracije aktivnosti ^{40}K, čije su se vrijednosti pokazale u rasponu (611,59-781,23) Bq kg^{-}1 veće kod svih analiziranih uzoraka zemljišta u poređenju sa onima dobijenim za ^{232}Th i ^{226}Ra, aktivnosti ^{137}Cs su generalno na nižem niovu i u rasponu (9,33-18,90) Bq kg^{-}1, sa najvećim aktivnostima na dubini od 5 cm, što je u velikoj mjeri u skladu sa podacima dobijenim iz literature.

Međutim, ^{40}K je jedini radionuklid kod kojeg je utvrđena značajna količina u uzorcima zemljišta i za koji se može reći da je na svim lokacijama na dubini od 5 cm pokazao visok stepen aktivnosti. Za sve ostale utvrđene radionuklide koji se prirodno pojavljuju smatra se da imaju nominalne koncentracije.

Na osnovu do sada dobijenih rezultata možemo zaključiti da uzorci zemljišta koji su sakupljeni u prirodnom okruženju posmatranih gradova ne ukazuju na pojačanu radioaktivnost, a koja bi mogla da predstavlja prijetnju za proizvodnju hrane i koja bi mogla da negativno utiče na ljudsko zdravlje. Koncentracije ovih radionuklida su na sličnom nivou sa raspoloživim podacima iz ostalih zemalja.

Ključne riječi: radioaktivnost; zemljište; radionuklidi; gama spektometrija; posebna aktivnost