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Radioactivity of the Agricultural Soil in Northern Province of Serbia, Vojvodina

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***Abstract*—**During the year 1999, Serbia (ex Yugoslavia) and their northern province, Vojvodina, has been bombarded. Because of that general public believe is that this region was contaminated by depleted uranium and that there is a potential contaminant of agricultural products due to soil radioactivity. This paper presents the repeated analysis of agricultural soil samples in Vojvodina. The same investigation was carried out during the year 2001, and it was concluded that, based on the gamma-spectrometric analysis of 50 soil samples taken from the region of Vojvodina, there haven't been registered any increase of radioactivity that could endanger the food production. We continue with the monitoring of this region. The comparison between those two sets of results is presented.

***Keywords*—**gamma spectrometry analysis, radioactivity of the agricultural soil.

I. Introduction

VOJVODINA is the northern province of Serbia. It is an agricultural region located in the Pannonian Plain of Central Europe. The economy of Vojvodina is largely based on developed food industry and fertile agricultural soil that make up 84% of its territory. About 70% of agricultural products is corn, 20% industrial herbs, and 10% other agricultural cultures. The soil of Vojvodina may contain radioactive contaminants from different sources. There are nuclear power plants in the South East Europe region that could contaminant this soil through the release of radionuclides into air and water. The application of phosphate fertilizers with high uranium concentration may also cause a gradual increase of the uranium series activity concentration in soil. During the year 1999, Serbia (ex Yugoslavia) and their northern province, Vojvodina, has been bombarded. Because of that general public believe is that this region was contaminated by depleted uranium and that there is a potential contaminant of agricultural products due to soil radioactivity.

The concentration of uranium and thorium in Earth's crust is in the range 1.1–10 ppm [1] for uranium and 10 ppm [2] for thorium. This corresponds to an activity concentration range of 13.5–123 Bq/kg for ^{238}U and $^{39.4}$ Bq/kg for ^{232}Th.

The anthropogenic radionuclides reach the soil by dispersion, where once absorbed represents a reservoir for the potential inhalation or ingestion by humans. The intensity of the processes of soil sorbtion/desorbtion, migration, retention and translocation is influenced by the nature of the given radionuclide, the type of soil and of crops grown on it, and the climatic conditions.During the year 2001, the radioactivity of the soil in Vojvodina was measured [3]. The main conclusion was that there haven't been presented any increase of radioactivity that could endanger the food production. The authors also concluded that there were no traces of depleted uranium in the treated soil samples. During the year 2010, Nuclear Physics Group from Novi Sad, continue with monitoring of radioactivity of agricultural soil in this region. In this paper we compared new results from this year with those results obtained during the year 2001. We used similar measurement technique, and the samples were collected from the same locations.

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II. Experimental Procedure

A. Equipment

Activity concentrations of radionuclides gamma emitters were determined by the method of low-level gamma spectrometry on actively and passively shielded germanium detectors with maximal background reduction. Two high resolution HPGe detectors were used. First of them, produced by CANBERRA has nominal efficiency of 36% and resolution of 1.79 keV. The detector was operated inside the 12 cm thick lead shield with 3 mm Cu inner layer. Second one, germanium detector made by ORTEC was extended range GMX type detector (10 keV-3 MeV) with nominal efficiency of 32% and resolution of 1.9 keV. The detector was shielded with the cylindrical lead shield of 12 cm wall thickness. Surrounding the lead shield, the five 0.5 m x 0.5 m x 0.05 m plastic veto detectors, produced by SCIONIX, were placed.

Veto plastic scintillators and Ge detector operate in anticoincidence mode and on that way all events that are simultaneously detected in any veto and Ge detector will be rejected.

The active shield reduces the integral background by factor 3 in the energy range from 50 to 2800 keV [4]. Through CANBERRA type pre-amplifiers and amplifiers spectra were channeled to multichannel analyzer MCA with two analog-digital converters of 8192 channels total memory. MCA was directly connected with PC in which measured spectra were stored and analyzed. The gamma spectra were acquired and analyzed using the Canberra Genie 2000 software.

The program calculates the activity concentration of an isotope from all prominent gamma lines after peaked background subtraction. All measurement uncertainties are presented at 95% confidence level. That means that probability of errors in repeated measurement of the same sample would be less than 5%.

A special procedure developed in the Novi Sad laboratory was used for the determination of the ^{238}U activity concentration from gamma-lines of the first progeny of this radionuclide, ^{234}Th [5].

B. Sample Treatment and Measurement

The samples were collected from the locations presented on the map on Figure 1. Fifty soil samples from Vojvodina were collected during the December 2010. From each location of an approximately 10×10 m area, 10 subsamples were collected, mixed and homogenized. The soil was sampled from the surface (0–2 cm).

Soil samples were dried at 105°C to constant mass. After that all mechanical contaminants, mainly small rocks and pieces of plant material were removed. Dried soil samples were mechanically fragmented and homogenized as fine powder. Prepared soil samples were packed in cylindrical measurement utensils 62 mm in height and 67 mm in diameter. Typical measurement time was 80 ks.

Fig. 1 The sampling locations in Vojvodina

III. Results and Discussion

Compared results of gamma spectrometry measurements of soil samples for the year 2001 and 2010, are presented in the Table 1. Activity concentrations of fission and corrosion products (except ^{137}Cs) were below detection limits, therefore in the final results only the activity concentrations of ^{137}Cs, the natural radioactive series of ^{238}U and ^{232}Th, and the natural radionuclide ^{40}K are presented.

TABLE I
Activity concentration of radionuclides in agricultural soil samples from Vojvodina

	Locations	Activity concentration of radionuclides, A [Bq/kg]									
		^{238}U		^{226}Ra		^{232}Th		^{40}K		^{137}Cs	
		2001	2010	2001	2010	2001	2010	2001	2010	2001	2010
1.	Becej	60±30	40±4	26.8±1.4	26.7±1.7	51.7±2.5	47.4±2.5	720±30	794±26	8.8±0.7	9.2±0.6
2.	Nadalj	54±17	36±4	44.1±1.9	33.8±2.5	52±3	41.5±2.6	553±29	556±19	13.3±1.2	7.7±0.5
3.	Srbobran	48±16	66±4	39.2±2.5	46.8±2.2	49.4±2.5	56.0±2.2	515±26	697±23	9.0±1.3	6.2±0.5
4.	Palic	24±9	9.4±1.7	19.9±1.8	9.7±1.3	23.5±2.6	11.7±0.5	310±20	238±10	8.3±0.9	6.3±0.4
5.	Coka	42±16	22.0±2.1	35.3±1.4	29.4±1.6	53.3±2.6	41±3	619±27	684±18	12.0±1.0	9.1±0.4
6.	Tornjos	58±17	78±11	43±3	41.9±2.3	54±3	45±5	492±27	560±50	10.0±1.1	3.8±0.5
7.	B.Novo Selo	35±11	25.3±2.0	30.2±1.9	29.4±1.3	36.7±2.3	30.9±2.9	464±21	554±15	6.0±0.7	4.45±0.23
8.	Srpski Miletic	64±29	< 20	40.5±2.7	40.5±2.1	57±3	39.2±2.7	580±30	452±24	6.4±0.8	3.6±0.3
9.	Orlovat	53±15	36±3	37.0±2.5	37.3±2.3	54.7±2.8	43.7±1.6	630±30	594±16	9.6±0.8	6.7±0.3
10.	Kikinda	43±18	<21	29.0±2.8	30.0±1.7	55.9±2.9	35±5	710±40	524±28	11.7±1.3	5.1±0.4

TABLE I
ACTIVITY CONCENTRATION OF RADIONUCLIDES IN AGRICULTURAL SOIL SAMPLES FROM VOJVODINA

	Locations	Activity concentration of radionuclides, A [Bq/kg]									
		^{238}U		^{226}Ra		^{232}Th		^{40}K		^{137}Cs	
		2001	2010	2001	2010	2001	2010	2001	2010	2001	2010
11.	Petrovaradin	60±18	66±4	42.2±2.0	47.9±2.5	62±3	64.4±2.5	590±30	705±23	13.8±1.6	6.7±0.5
12.	Kac	56±27	35±3	44.1±2.6	28.7±2.6	48±3	32±4	600±30	530±20	8.1±1.1	8.0±0.6
13.	Idvor	41±19	25±3	21.3±2.4	34.0±2.8	34±4	47.2±2.3	350±26	703±22	7.2±1.1	10.2±0.6
14.	Boka	53±18	44±4	44.2±2.1	34.1±1.7	64±4	43.4±2.7	550±30	633±21	10.3±1.1	10.3±0.6
15.	Sanad	49±14	54±4	34.9±2.0	35±4	51±3	61.1±1.4	640±40	1000±30	9.3±1.2	4.4±0.5
16.	Kula, Lipar	56±23	28.3±2.6	43.2±2.3	38.5±2.4	54±3	45.8±2.9	493±27	552±15	7.8±1.3	6.9±0.3
17.	Parage	66±19	38.5±2.9	45.5±2.9	39.3±1.3	57±4	45.1±1.2	560±30	588±21	9.4±1.4	5.6±0.5
18.	Rivica	63±16	80±10	45.2±2.8	43.4±2.8	63±3	48±5	560±30	520±40	8.5±0.7	5.5±0.5
19.	Visnjicevo	55±17	< 22	51.0±2.1	34.3±1.9	63±3	35±11	610±30	507±27	8.1±1.1	12.2±0.7
20.	Maglic	39±15	35±4	40.6±2.5	36.4±2.0	52.3±2.8	38±5	513±28	465±25	5.7±0.9	3.04±0.29
11.	Sid	69±16	< 21	44.4±2.2	40.2±2.1	59±3	44±5	567±28	521±27	7.5±0.7	3.3±0.3
12.	Vrsac	50±20	74±3	31.9±2.2	49.1±2.6	59±4	70.5±2.3	580±30	701±24	12.6±1.4	5.0±0.6
13.	Crepaja	44±24	67±10	36.4±2.3	43.1±2.2	50.7±2.5	42±5	480±30	500±40	17.5±1.4	9.9±0.7
14.	Gakovo	55±16	47±3	41.6±2.5	38.6±1.1	52±3	44.2±1.1	475±25	497±18	7.0±0.9	2.3±0.4
15.	Zrenjanin	49±23	25.3±2.5	40.4±1.8	37.0±1.6	50.1±2.4	48±3	526±25	677±8	8.5±1.1	5.6±0.3
16.	Padina	55±22	< 21	41.0±2.4	41±3	55±3	41±5	534±29	492±26	12.3±1.1	6.7±0.5
17.	Deliblato	51±14	16.9±1.6	41.5±2.7	20.7±1.2	54.3±2.9	24.3±2.7	501±23	399±11	28.2±1.9	20.4±0.5
18.	R. Sancevi	52±16	73±10	40.5±2.6	39.9±2.5	54±3	43±4	610±30	560±50	6.9±1.4	5.1±0.5
19.	Bogojevo	42±17	17.2±1.6	37±3	23.5±1.8	48.2±2.8	24.1±2.7	543±28	428±12	9.2±1.0	4.49±0.23
20.	Rusko Selo	53±14	30±3	34.9±1.5	25.1±2.3	54±3	35.4±1.8	730±40	621±20	19.7±1.2	11.9±0.7
21.	Morovic	56±18	72±10	50.9±1.8	36.2±2.8	59±3	48±6	571±26	520±40	7.8±0.7	12.1±0.8

TABLE I
ACTIVITY CONCENTRATION OF RADIONUCLIDES IN AGRICULTURAL SOIL SAMPLES FROM VOJVODINA

	Locations	Activity concentration of radionuclides, A [Bq/kg]									
		^{238}U		^{226}Ra		^{232}Th		^{40}K		^{137}Cs	
		2001	2010	2001	2010	2001	2010	2001	2010	2001	2010
22.	Ruma, Irig	50±16	68±4	50.4±2.0	47.0±2.1	60±3	64.8±2.0	557±29	758±24	7.7±0.9	7.9±0.6
23.	A. Santic	60±17	< 21	46.6±2.4	39±4	54.4±2.7	37±3	470±30	380±21	6.7±0.7	4.4±0.4
24.	Bavaniste	59±17	37±3	43±4	37.3±2.7	55.2±2.9	45.5±2.1	550±30	623±20	55±3	42.6±1.2
25.	Pecinci, Popinci	56±16	38±4	39.4±2.9	34.1±1.6	56±3	47±3	534±28	569±19	11.5±1.6	9.9±0.6
26.	R. Krstur	56±15	26.0±2.3	48.6±2.0	35.6±1.4	53.7±2.6	41.5±2.8	523±24	581±16	7.7±0.7	5.60±0.27
27.	Zednik	49±22	< 20	40.1±2.6	36.6±1.9	50.5±2.6	35.9±2.3	488±28	392±21	11.1±1.5	5.5±0.4
28.	Horgos	31±9	24±5	19.7±1.0	12.4±0.8	22.0±1.5	14.6±1.2	238±13	249±21	1.1±0.3	5.6±0.4
29.	Ilandza	57±20	< 22	42.3±2.4	42.3±2.3	57±3	44±3	550±40	521±28	18.4±1.5	12.4±0.7
30.	Kozjak	52±16	43±7	40.1±2.7	41±3	50.1±2.7	38.7±2.9	500±30	480±40	15.5±1.3	11.0±0.6
31.	Zabalj	45±17	52±5	41.8±2.4	36.7±2.5	59±3	47.7±2.0	640±30	658±22	10.5±1.0	9.0±0.6
32.	Vrsacki Ritovi	30±24	53±4	26.2±1.7	33.1±2.9	54.8±2.8	56.7±2.6	551±23	745±25	48.7±2.4	25.8±1.0
33.	Begejci	56±18	61±4	50.9±1.8	45.1±2.6	59±3	57.9±1.4	571±26	786±26	7.8±0.7	6.7±0.6
34.	B. Arandjelovo	50±16	70±10	50.4±2.0	30.0±1.8	60±3	41±5	557±29	590±50	7.7±0.9	6.5±0.6
35.	Torda	55±16	< 22	40.3±2.0	37±3	59±3	46±3	660±30	590±30	15.5±1.3	10.1±0.6
36.	Kumane	49±22	59±9	37.9±2.0	31.1±1.7	55.0±2.7	45±4	720±30	640±50	6.8±0.6	7.8±0.6
37.	Donji Tovarnik	50±23	76±10	40±3	41.7±2.1	62±3	50±10	600±30	610±50	9.4±1.1	7.4±0.6
38.	Obedska Bara	72±21	31.1±2.8	43.3±2.5	24.9±2.7	62±3	34.8±1.0	630±30	479±19	11.9±1.1	18.4±0.9
39.	Indjija	57±23	< 20	44±3	38.6±2.0	59±3	35±13	580±30	488±26	6.7±0.9	4.5±0.4
40.	Sr.Mitrovica	49±15	73±9	39.9±2.4	35.3±2.1	55±3	42.3±2.9	536±24	530±40	5.9±0.6	5.0±0.5

Distributions of ^{238}U, ^{232}Th, ^{40}K and ^{226}Ra are shown on the Figures 2 – 5.

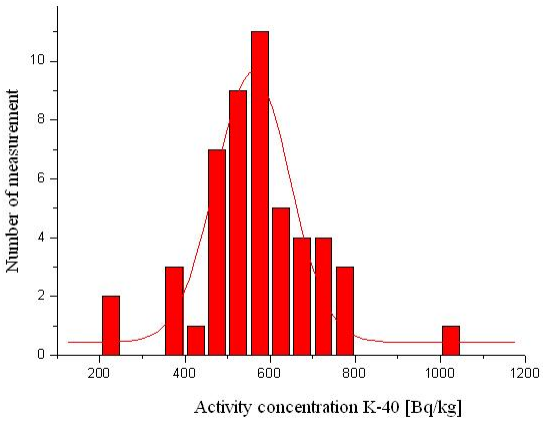

Fig. 2 Distribution of activity concentrations of ^{40}K

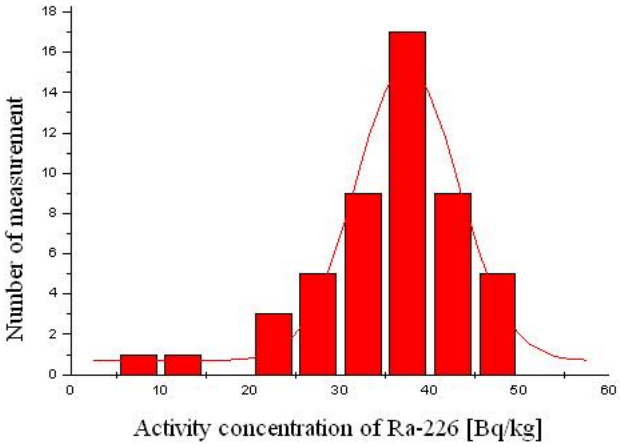

Fig. 3 Distribution of activity concentrations of ^{226}Ra

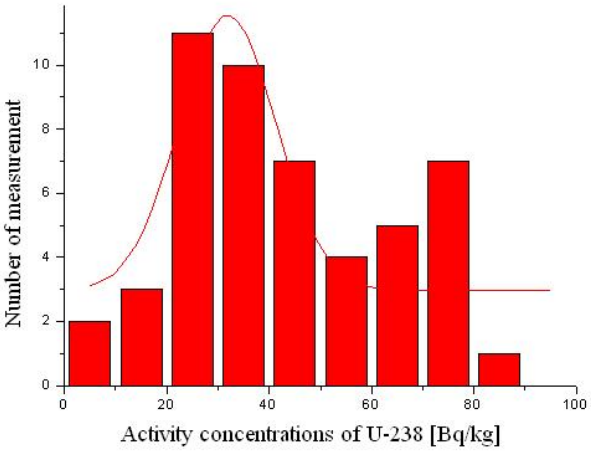

Fig. 4 Distribution of activity concentrations of ^{238}U

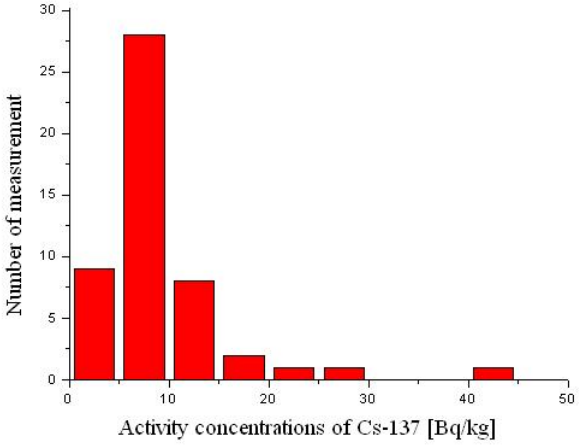

Fig. 5 Distribution of activity concentrations of ^{137}Cs

IV. Conclusion

The activity concentration of the natural radioactive series of ^{238}U, ^{232}Th, ^{226}Ra and the natural radionuclide ^{40}K are at the normal environmental levels. The radionuclide ^{137}Cs was identified in all soil samples. The distribution of ^{137}Cs is skewed which is typical for a man made contaminant. This radionuclide originates from nuclear weapons test fall-out and, after 1986, mostly from the accident of the nuclear power plant ‘‘Lenin’’ in Chernobyl. Due to the 30 year half-life of this radionuclide, it will be relocated, washed out and redistributed. However it will be present for a long time in the Vojvodina ecosystem. If we compare ^{137}Cs activity concentration results with previous measurements, we note a slight decrease after 2001. Only in locations 1, 13, 19, 24, 38, 47, 48, the recent values of activity concentrations of ^{137}Cs are higher than in 2001, probably due to soil deposition.

Since the ^{238}U activity concentration in all samples is at the natural environmental level, and the $^{238}U/^{226}Ra$ ratio is not

substantially changed, one can conclude that in the measured samples there is no indication of depleted uranium presence. The activity concentration of the natural radioactive series of ^{232}Th, and the natural radionuclide ^{40}K are also at the normal environmental levels.

If we compare the ^{238}U activity concentration results with previous measurements, we note a slight discrepancy probably due to use of phosphate fertilizers. Taking into account the transfer factors of present radionuclides to plants, the measured activity concentrations of radionuclides in Vojvodina agricultural soil should not endanger the health safety of the produced food.

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