RADIONUCLIDES IN AGRICULTURAL SOIL IN VOJVODINA REGION

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Abstract
The soil of Vojvodina is subject to radioactive contamination from a number of sources, reactors of nuclear power plants in the South Europe region and use of phosphate fertilizers with high uranium concentration. The results of radiological control in agricultural soil in Vojvodina region are presented in this paper. Based on gamma-spectrometric analysis of 50 soil samples taken from the region of Vojvodina one can conclude that there is no increase of radioactivity that could endanger the food production.

Introduction

The results of measurements of radionuclides activity concentration in the agricultural soil in the Vojvodina region are presented in this paper*.* The soil of Vojvodina may contain to radioactive contaminants from a number of sources. First of all, these are the nuclear power plants in the South East Europe region that could contaminate this region through the release of radionuclides into air and water. The use of phosphate fertilizers with high uranium concentration may also cause a gradual increase of the uranium series activity concentration in soil (1).
Generally, the all soil samples taken from all location do not show increasing radioactivity which will be endanger the food production. The measured values of ^{137}Cs activity, using the transfer factor of this isotope into plants, should not endanger the health safety of food production.

Methods

The soil samples were dried at 105°C to constant mass and transferred to sample holders. Gamma-spectrometric measurements were performed with an high-resolution HPGe gamma-spectrometer made by ORTEC. The nominal detector efficiency exceeds 36%, while the resolution is less than 1.9 keV. The detector has an increased energy range of measurement (GMX-type) such that it can detect also low-energy γ- and X-radiation. The metallic parts of the detector were made of materials tested for high radiopurity. The detector was placed in a special low background protection chamber with iron walls 25 cm thick. The chamber is made of pre WW II cast iron, so that it does not contain admixtures of man-made radioactivity thereby reducing the background radiation level for about 1000 times. The spectra were led through the preamplifier-amplifier chain (the latter of CANBERRA-type) to the CANBERRA Series 35+ multi channel analyzer with two analog-to-digital converters, and with a memory containing 8192 channels. The multi channel analyzer is directly connected to a personal computer where the spectra were processed and stored. A modified version of the SAMPO program was used to process the spectra, in such a way that, besides the identified gamma-lines, it always presented spectral intensities of 20 selected isotopes. The samples were measured in cylindrical geometry, placed in sample containers with 67 mm diameter and 62 mm height. The detection efficiency for this geometry was determined with primary calibration point sources made by AMERSHAM, with calibrated voluminous sources made by NBS and OMH, as well as with a phosphate-ore sample of known activity concentration. The consistency of the calibration results was checked with a modified version of the SOLANG computer program. Typical measurement time was 80 ks. The measurement uncertainties were presented at the 95% confidence level, what means that the probability for obtaining a result laying outside the presented limits in a repeated measurement of the same sample is less than 5%. Activity concentrations of fission and corrosion products (except ^{137}Cs) were below the 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.

A special procedure developed in the Novi Sad laboratory enables the determination of ^{238}U activity concentration from gamma-lines of the first progeny of this radionuclide, ^{234}Th. Besides the ^{238}U activity concentration determined in this way, the activity concentrations of the ^{226}Ra member of the uranium series are also presented. The comparison of these two measurements provides indication on the presence of depleted uranium because in materials contaminated with depleted uranium the equilibrium ratio of uranium to radium is substantially disturbed (2).

Results

Activity concentrations of all measured radionuclides, except ^{137}Cs, are below the detection limit. The radionuclide ^{137}Cs is present in all soil samples. This radionuclide originates from the accident of the nuclear power plant 'Lenin' in Chernobyl in 1986. Due to the long half-life of this radionuclide of 30 y, it will be relocated, washed out and redistributed, but it will be present for a long time in the Vojvodina ecosystem. The large standard deviation and the large difference between the minimum and maximum ^{137}Cs activity concentrations show typical features of a man-made contaminant (3).
In Table 1 are present the measured activity concentration for radionuclides in soil under corn and wheat.

Table 1. Activity concentration of radionuclides in soil under corn and wheat

Radio nuclide	Soil 1 Elan P1	Soil 2 Elan P2	Soil 3 Elan P3	Soil 4 ElanT4	Soil 5 Elan T-939	Soil 6 Skorenovac P1	Soil 7 Skorenovac 13	Soil 8 Skorenovac 1a
				A[Bq/kg]				
^{75}Se	<0.17	<0.6	<0.6	<0.4	<0.29	<0.23	<0.18	<0.24
^{144}Ce	<1.7	<2.3	<2.1	<2.3	<2.0	<1.3	<1.6	<3.0
^{141}Ce	<0.29	<0.8	<1.0	<0.7	<0.5	<0.10	<0.5	<0.6
^{125}Sb	<0.9	<0.9	<1.0	<0.5	<1.6	<0.8	<0.6	<0.6
^{7}Be	<2.3	<3	<3	<4	<4	3.0±2.5	<4.4	<2.5
^{103}Ru	<0.3	<0.6	<0.4	<0.3	<0.6	<0.26	<0.3	<0.57
^{134}Cs	<0.5	<0.5	<0.7	<0.17	<0.10	<0.21	<0.16	<0.25
^{124}Sb	<0.27	<0.9	<0.4	<0.7	<0.6	<0.22	<0.25	<0.16
^{106}Ru	<2.8	<3	<2.9	<9	<7	<5	<2.5	<2.3
^{110m}Ag	<0.43	<0.14	<0.16	<0.28	<0.02	<0.25	<0.06	<0.34
^{137}Cs	7.8±1.2	7.9±1.0	7.9±1.0	10.6±1.5	8.5±1.0	54±4	30.7±1.5	47.6±1.9
^{95}Zr	<0.4	<0.9	<0.4	<0.5	<0.7	<0.27	<0.5	<0.6
^{95}Nb	<0.5	<0.6	<0.5	<0.4	<0.7	<0.3	<0.6	<0.09
^{58}Co	<0.4	<0.4	<0.6	<0.4	<0.4	<0.3	<0.26	<0.23
^{160}Tb	<1.6	<1.9	<1.3	<2.2	<2.9	<1.2	<1.5	<1.5
^{60}Co	<0.28	<0.4±0.4	<1.0	<0.29	<0.4	<0.29	<0.22	<0.21
^{238}U	37±17	50±26	84±28	70±40	40±30	40±20	80±29	45±17
^{226}Ra	40.4±1.7	41.5±1.9	39.2±2.2	49±3	46±3	30±3	31.9±1.6	28.5±1.1
^{232}Th	46.7±2.5	46.1±2.5	43.6±2.6	57±3	57±6	49±5	54.7±2.5	44.0±2.2
^{40}K	510±30	500±30	480±40	571±28	588±29	601±27	660±30	513±26

As can be conclude from Table 1., the activity concentration of all radionuclides, except from ^{137}Cs are under detection limit. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclide in soil under corn and wheat are presented in Table 1a.
The highest values of ^{137}Cs activity concentrations are in the location: Skorenovac P1, 13 and 1a. However, all measured activity concentrations are convenience for growing of corn and wheat.
Measured values of acitvity concentration of radionuclides for soil under soya bean and sunflower are presented in Table 2.

Tabele 1a. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclides in soil under corn and wheat

Radionuclide	$\bar{A}_s$ [Bq/kg]	$\sigma\ \bar{A}_s$ [Bq/kg]	A_s (min) [Bq/kg]	A_s(max) [Bq/kg]
^{137}Cs	22	20	7.8	54
^{238}U	56	19	37	84
^{226}Ra	38	8	28.5	49
^{232}Th	50	6	43.6	57
^{40}K	553	62	480	601

Table 2. Activity concentration of radionuclides in soil under soya bean and sunflower

Radio nuclide	Soil 1 Karavuko vo P-1	Soil 2 Karavuko vo P-2	Soil 3 Bečej P1	Soil 4 Bečej P2	Soil 5 Karavukovo peskuša	Soil 6 Karavukovo kanalčići	Soil 7 Bečej T-134	Soil 8 Bečej T-140
			A[Bq/kg]					
^{75}Se	<0.20	<0.18	<0.4	<0.5	<0.19	<0.33	<0.25	<0.21
^{144}Ce	<1.8	<1.3	<2.3	<1.9	<1.6	<1.4	<1.5	<1.2
^{141}Ce	<0.16	<0.3	<0.4	<0.7	<0.5	<10	<0.4	<0.50
^{125}Sb	<0.8	<1.1	<0.9	<1.1	<1.31	<0.9	<0.6	<0.7
^{7}Be	<3.0	<2.2	<3	<5	<5	<2.5	<3.6	<1.8
^{103}Ru	<0.21	<0.37	<0.7	<0.3	<0.5	<0.47	<0.24	<0.19
^{134}Cs	<0.4	<0.25	<0.4	<1.1	<0.4	<0.22	<0.07	<0.7
^{124}Sb	<0.30	<2.5	<0.4	<0.5	<0.8	<0.4	<0.36	<0.4
^{106}Ru	<2.1	<2.9	<3	<3	<3	<2.1	<2.6	<2.7
^{110m}Ag	<0.29	<0.05	<0.4	<0.34	<0.46	<0.16	<0.28	<0.43
^{137}Cs	4.2±0.5	5.0±1.2	8.8±1.0	11.1±1.4	5.3±0.6	5.1±1.4	10.9±0.9	10.9±1.0
^{95}Zr	<0.7	<0.4	<1.1	<0.4	<0.4	<0.8	<0.	<0.5
^{95}Nb	<0.24	<0.08	<0.7	<0.08	<0.5	<0.37	<0.34	<0.27
^{58}Co	<0.27	<0.4	<0.4	<0.4	<0.3	<0.23	<0.22	<0.22
^{160}Tb	<0.7	<1.0	<2.1	<1.6	<1.2	<0.8	<0.7	<0.7
^{60}Co	<0.20	<0.21	<0.3	<0.27	<0.25	<0.19	<0.20	<0.19
^{238}U	42±15	34±10	53±25	36±21	70±30	40±21	57±20	54±20
^{226}Ra	38.3±2.7	42±4	40.5±2.4	47±4	39.9±2.0	38±4	41.5±1.4	46.5±2.5
^{232}Th	40.8±2.2	47.3±2.9	46±3	48±4	38.5±2.0	41±3	44.1±2.1	53±5
^{40}K	577±29	581±24	520±40	560±40	560±40	576±25	511±27	586±23

As can conclude from Table 2., the activity concentration of all radionuclides are under detection limit. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclide in soil with soya bean and sunflower are presented in Table 2a.

Tabele 2a. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclides in soil under soya bean and sunflower

radionuclide	$\bar{A}_s$ [Bq/kg]	$\sigma\ \bar{A}_s$ [Bq/kg]	A_s (min) [Bq/kg]	A_s(max) [Bq/kg]
^{137}Cs	7	3	4.2	11.1
^{238}U	48	12	34	70
^{226}Ra	42	3	38	46.5
^{232}Th	45	5	38.5	47.3
^{40}K	559	28	511	586

All location from Table 2, from the radioactivity wiev, are convenience for growing of soya bean and sunflower.

Measured values of acitvity concentration of radionuclides for soil under vegetables are presented in Table 3.

Table 3. Activity concentration in soil under vegetable

Radio nuclid e	Soil 1-BAG B. Gradište T21	Soil 2 BAG B. Gradište -T20	Soil 3-BAG B. Gradište-T5/1	Soil 4-BAG B. Gradište T5/2	Soil 5-BAG B. Gradište T4	Soil 6-BAG B. Gradište T-6	Soil 7-BAG B. Gradište T-20	Soil 8-BAG B. Gradište T-9
				A[Bq/kg]				
^{75}Se	<0.27	<0.23	<0.24	<0.13	<0.20	<0.4	<0.30	<0.21
^{144}Ce	<1.5	<1.6	<1.3	<2.0	<2.0	<1.8	<1.5	<1.3
^{141}Ce	<0.3	<0.20	<0.7	<0.5	<0.5	<0.9	<0.4	<0.5
^{125}Sb	<0.7	<0.9	<0.7	<0.7	<1.8	<1.1	<1.0	<0.8
^{7}Be	<2.9	<3.5	<3.4	<2.3	<5	<2.5	<2.4	<2.9
^{103}Ru	<0.25	<0.32	<0.22	<0.29	<0.4	<0.3	<0.28	<0.24
^{134}Cs	<0.16	<0.6	<0.3	<0.22	<0.6	<0.6	<0.22	<0.8
^{124}Sb	<0.20	<0.4	<0.36	<0.49	<0.5	<1.2	<0.33	<0.6
^{106}Ru	<2.0	<2.0	<2.1	<2.5	<4	<4	<2.3	<2.2
^{110m}Ag	<0.29	<0.3	<0.18	<0.06	<0.47	<0.3	<0.21	<0.13
^{137}Cs	8.9±0.8	12.6±1.2	8.8±1.3	8.2±0.8	8.6±1.0	11.3±1.7	8.0±0.7	8.0±0.6
^{95}Zr	<0.6	<0.5	<0.3	<0.4	<0.4	<0.8	<0.5	<0.4
^{95}Nb	<0.3	<0.45	<0.47	<0.4	<1.0	<0.5	<1.7	<0.33
^{58}Co	<0.28	<0.5	<0.4	<0.29	<0.5	<0.5	<0.55	<0.36
^{160}Tb	<1.2	<0.6	<1.1	<0.8	<0.5	<1.3	<0.8	<1.5
^{60}Co	<0.23	<0.21	<0.19	<0.28	<0.3	<0.5	<0.21	<0.5
^{238}U	55±19	50±17	49±19	70±50	70±30	59±25	71±22	56±14
^{226}Ra	37.1±1.3	46±3	45±3	36.5±1.4	35.9±1.6	44.9±2.4	39.9±1.3	45±3
^{232}Th	47.1±2.1	57±3	51±5	47.5±2.2	46.5±2.5	52±3	47.1±2.2	58±4
^{40}K	506±26	612±25	569±23	531±29	510±30	630±30	527±27	610±30

The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclide in soil under vegetable are presented in Table 3a.

Table 3a. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclides in soil under vegetable

radionuclide	$\bar{A}_s$ [Bq/kg]	$\sigma\ \bar{A}_s$ [Bq/kg]	A_s (min) [Bq/kg]	A_s(max) [Bq/kg]
^{137}Cs	9	2	8.9	126
^{238}U	54	14	49	71
^{226}Ra	41	5	35.9	46
^{232}Th	50	5	46.5	58
^{40}K	565	58	506	630

All location from Table 3, from the radioactivity wiev, are convenience for growing of vegetable.

Measured values of acitvity concentration of radionuclides for soil potatoes are presented in Table 4. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclide in soil under potatoes are given in Table 4a.
All location from Tables 4 and 4a, from the radioactivity wiev, are convenience for growing of potatoe.

Conclusion

The fission product from Chernobyl ^{137}Cs is present in all soil samples. No traces of other produced radionuclides are found. The ratio of ^{238}U and ^{226}Ra is approximately constant in all samples. Activity concentration of ^{238}U is in the common level in all samples, it could be concluded that there is no depleted uranium in soil. Activity concentration of natural radioactive chain ^{232}Th and ^{40}K are in similar that the values in middle Europe. Generally, it could be concluded that there is no increase in the radioactivity in the location of agricultural soil.

Table 4. Activity concentration in soil under potatoes

Radionuclide	Soil 2 Maglić T-10	Soil 4 Maglić T-9	Soil 5 Maglić T-12	Soil 6 Maglić T-20	Soil 7 Maglić T-20/2	Soil 8 Maglić T-1	Soil 9 Dugalić	Soil 10 Dugalić	Soil 11 Dugalić
					A[Bq/kg]				
^{75}Se	<0.35	<0.19	<0.6	<0.4	<0.23	<0.21	<0.19	<0.19	<0.50
^{144}Ce	<3.8	<1.9	<3.0	<2.2	<1.5	<1.4	<1.9	<1.2	<1.9
^{141}Ce	<1.1	<0.6	<0.16	<0.25	<0.7	<0.7	<0.3	<0.4	<0.5
^{125}Sb	<2.1	<0.5	<1.4	<2.7	<0.5	<1.7	<1.4	<1.4	<1.9
^{7}Be	<3	<1.8	<3	<4	<2.3	<1.7	<2.5	<2.2	<3.5
^{103}Ru	<0.4	<0.21	<0.4	<0.7	<0.28	<0.21	<0.47	<0.24	<0.5
^{134}Cs	<0.5	<0.8	<0.4	<1.6	<0.3	<0.6	<0.18	<0.09	<0.5
^{124}Sb	<0.7	<0.52	<0.7	<1.0	<0.26	<0.5	<0.17	<0.4	<0.15
^{106}Ru	<5	<2.6	<6	<3	<4	<2.2	<1.9	<2.3	<5.3
^{110m}Ag	<0.5	<0.21	<0.02	<0.3	<0.34	<0.7	<0.25	<0.15	<0.09
^{137}Cs	6.9±0.9	6.3±1.3	6.0±1.7	4.5±1.3	6.6±0.6	7.8±1.4	54.3±2.2	79±4	108±5
^{95}Zr	<0.8	<0.5	<1.1	<0.7	<0.6	<7	<0.30	<1.0	<0.3
^{95}Nb	<0.7	<0.48	<1.0	<0.5	<0.27	<0.5	<0.4	<0.5	<0.4
^{58}Co	<0.3	<0.35	<0.5	<0.6	<0.24	<0.23	<0.18	<0.54	<0.25
^{160}Tb	<1.4	<0.9	<2.2	<1.8	<0.5	<1.0	<0.6	<1.3	<1.3
^{60}Co	<0.3	<0.21	<0.5	<0.28	<0.35	<0.23	<0.16	<0.18	<0.23
^{238}U	40±15	53±15	70±30	60±30	60±21	49±20	15±10	31±15	53±14
^{226}Ra	39.4±1.8	48±3	39.6±2.0	49.3±2.8	38.3±1.5	46±3	4.4±0.7	23.8±2.9	46±4
^{232}Th	50.6±2.6	61±3	49.7±2.7	57±5	51.8±2.4	60±3	6.9±0.8	17.1±1.8	67±5
^{40}K	530±40	655±28	570±40	581±28	568±29	621±28	82±10	230±13	980±40

Table 4a. The average values (with standard deviations) and the minimum and maximum values of activity concentrations of detected radionuclides in soil under potatoes.

radionuclide	$\bar{A}_s$ [Bq/kg]	$\sigma \bar{A}_s$ [Bq/kg]	A_s (min) [Bq/kg]	A_s(max) [Bq/kg]
^{137}Cs	27	37	4.5	108
^{238}U	48	15	15	70
^{226}Ra	38	13	4.4	49.3
^{232}Th	48	19	6.9	60
^{40}K	540	229	82	980

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