RADIONUCLIDES IN THE SOIL OF VOJVODINA NATURE RESERVES

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Abstract

Protected environmental areas should preserve the undisturbed natural conditions for all living species. However human activities have impact also on these areas. Contamination with long-lived produced and natural radionuclides is one of the factors which can disturb the historical ambient. In this work the radionuclide content of the soil in about 20 nature reserves areas will be presented and discussed.

Introduction

The soil of Vojvodina may contain 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 the water. The use of phosphate fertilizers with high uranium concentration may also cause a gradual increase of the uranium series activity concentration in soil. 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. Besides the daughters of natural radioactive series, the long-lived natural isotope ^{40}K is also present in the soil.
The anthropogenic radionuclides reach the soil by dispersion. Due to its absorbing properties the soil represents a permanent reservoir for the potential inhalation or ingestion by humans. The intensity of the radioactivity transport processes in soil (sorbtion/desorbtion, migration, retention and translocation) is influenced by the nature of the given radionuclide, as well as by the type of soil and type of crops grown on it.

The activity concentrations of natural radionuclides in the rocks are well known (Table 1) [3,4]. The soil of Vojvodina, in its inorganic component contents silicates originating from the decay of granite rocks with high concentration of thorium and uranium. Due to different organic and inorganic composition of soil and different structure, wide range of radionuclide concentrations can be expected.

Table 1 The activity concentrations of natural radionuclides in the rocks

Radionuclide	**Rocks**		
	basalt	sienna	granite
^{40}K [B/kg]	210	1400	1290
^{232}Th [B/kg]	6.5	69.2	87.5
^{238}U [B/kg]	5.3	102.0	59.7

Methods

The soil samples were collected from the surface. The average mass of samples was 1kg. The soil samples were dried at 105^{o}C to constant mass and transferred to sample holders. Gamma spectrometric measurements were performed with two high resolution HPGe gamma spectrometers.The first one, the GMX type, made by ORTEC with nominal efficiency exceeding 36% and reslution less than 1.9 keV, was placed in a special low background schielding chamber with lead walls 12 cm thick. The second CANBERRA HPGe detector with nominal efficiency 22%, was placed in another schielding chamber with iron walls 25 cm thick. The pulses were led through the preamplifirier-amplifier chain to CANBERRA Ser. 35+ multi channel analyzer with two analog-to-digital converters, and with a memory containing 8192 channels. The multi channel analyzer was directly connected to a personal computer where the spectra were processed and stored. A modified version of the SAMPO program was used to evaluate 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 diametar 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 the 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 80ks. The measurement uncertaintieswere presented at the 95% confidence level. 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 Novi Sad laboratory [5] enables the determination of the ^{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.

Results

Measured soil samples were collected from several locations in Nature Reserves of Vojvodina (Fig.1). Measurements of radioactivity were performed in Laboratory of Nuclear Physics, Department of Physics in Novi Sad. The results of these measurements are presented in Table 1.

Fig. 1 The map of Vojvodina province with the locations of Nature Reserves

Table 1. The activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil samples

		ACTIVITY CONCENTRATION [Bq/kg]				
Nature Reserve sample	GPS coordinates	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Fruška Gora 1	X 7410883 Y 5002232	62±17	48±5	56.3±2.8	700±30	25.6±1.7
Fruška Gora 2	X 7401878 Y 5002125	50±40	34.5±1.5	38.6±2.3	379±25	11.4±1.0
Fruška Gora 3	X 7422759 Y 5003169	28±12	32±4	40.4±2.4	450±30	24±3
Fruška Gora 4	X 7403760 Y 5000841	<60	50.3±2.6	56±3	530±40	14.2±2.6
Fruška Gora 5	X 7399096 Y 5002069	70±25	44±3	70±4	880±40	5.5±1.2
Fruška Gora 6	X 7399134 Y 4996643	48±23	44.0±1.9	46.7±2.3	450±30	15.8±1.2
Fruška Gora 7	X 7400591 Y 5007059	40±11	28.0±1.7	25.9±2.0	328±17	<0.6
Obedska bara 1	X 7421196 Y 4955746	65±24	41.8±1.9	46.4±2.2	540±40	22.2±1.4
Obedska bara 2	X 7424009 Y 4950780	70±26	37.6±1.6	44.3±2.3	590±30	36.5±1.9
Obedska bara 3	X 7419620 Y 4953930	44±28	33.9±1.5	38.2±2.0	436±27	45.6±2.4
Carska bara 1	X 7454843 Y 5015573	69±19	41±3	57.9±2.8	730±40	27.0±1.6
Carska bara 2	X 7453698 Y 5014199	50±30	26.6±1.3	24.2±1.4	323±22	17.3±1.2
Carska bara 3	X 7453923 Y 5015098	22±11	25.1±2.3	26.9±1.7	387±17	18.0±1.2
Ludoš 1	X 7407813 Y 5107245	<40	22.8±1.5	22.3±1.3	255±17	18.3±1.1
Ludoš 2	X 7409974 Y 5107438	18±7	17.1±1.3	19.1±1.5	282±18	9.9±1.2
Deliblatska peščara 1	X 7510918 Y 4977985	50±30	23.9±1.7	23.4±1.8	298±28	18.7±1.7
Deliblatska peščara 2	X 7510918 Y 4977985	24±18	26.0±2.9	32.2±1.7	416±19	17.5±1.2
Karađorđevo	X 7361700 Y 5023964	52±21	34.7±1.6	39.9±2.2	520±30	13.4±1.6
Zasavica 1	X 7384286 Y 4981125	40±30	39±3	45±4	590±30	9.1±1.5
Zasavica 2	X 7377680 Y 4976980	38±26	26.1±2.7	32.5±2.4	450±40	25.2±2.6
Gornje Podunavlje 1	X 7339862 Y 5076620	46±15	44±4	50±3	640±30	15.6±1.6
Gornje Podunavlje 2	X 7338121 Y 5049368	90±40	34±3	34.4±2.3	500±40	18.1±1.7
Velike droplje	X 7445919 Y 5084728	47±15	31±3	49±3	680±40	16.4±2.2
Selevenjska pustara	X 7416730 Y 5113091	<30	19.6±1.4	17.7±1.4	196±23	12.3±1.2
Slano Kopovo 1	X 7439598 Y 5053003	<24	26.8±1.4	29.0±1.8	460±30	11.0±1.2
Slano Kopovo 2	X 7439403 Y 5053194	29±12	29.0±1.6	34.8±2.0	540±30	5.9±1.4
Vršačke planine 1	X 7529538 Y 4994682	53±18	36±4	60±3	650±30	31.9±2.3
Vršačke planine 2	X 7526112 Y 4997801	40±30	37±5	57±4	780±40	33.9±2.4
Begečka jama	X 7390846 Y 5009928	42±25	35.0±2.9	34.8±2.1	500±27	12.3±1.6
Palić	X 7403687 Y 5106708	<30	27.3±1.5	26.8±1.9	280±30	15.3±2.0
Ponjavica	X 7485156 Y 4953101	27±16	31.3±1.8	30.9±1.7	500±30	6.3±0.9
Titelski breg	X 7439108 Y 5009002	38±17	53±3	56±3	511±29	15.3±1.7
Horgoška peščara	X 7395283 Y 5113382	38±20	25.9±1.8	20.1±1.6	263±24	10.2±1.2
Koviljsko-Petrovaradinski rit 1	X 7427422 Y 5005664	<35	30.6±1.6	24.2±1.4	355±25	14.2±1.8
Koviljsko-Petrovaradinski rit 2	X 7413940 Y 5009807	44±20	43±4	50±3	590±30	15.9±1.4
Biserno ostrvo	X 7427522 Y 5037884	59±23	30.8±1.7	38.5±2.6	580±40	27.1±1.8
Jegrička	X 7438008 Y 5028941	47±17	32.9±2.8	53±4	760±40	20.3±1.8

Discussion

The activity concentrations of measured radionuclides were found at natural environmental level. The radionuclide ^{137}Cs was identified in all soil samples.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. This radionuclide originates from the accident of the nuclear power plant 'Lenin' in Chernobyl in 1986. Due to 30 years 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.
The results of this investigation could be compared with the previous measurement of agricultural soil (Table 2). As can be noticed, the average concentrations of detected radionuclides in soil samples from Nature Reserves are lower than concentrations in samples of agricultural soil, except the concentration of ^{137}Cs. We assume that this radionuclide remain on the surface layer because soil in Nature Reserves haven't been ploughed and treated like the agricultural soil.
Because 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.

Table 2. The average $\bar{A}_{av}$, minimal $A_{(min)}$ and maximal $A_{(max)}$ concentration of radionuclides in agricultural soil and soil from natural reserves

	agricultural soil			nature reserves		
radionuclide	$\bar{A}_{av}$ [Bq/kg]	$A_{(min)}$ [Bq/kg]	$A_{(max)}$ [Bq/kg]	$\bar{A}_{av}$ [Bq/kg]	$A_{(min)}$ [Bq/kg]	$A_{(max)}$ [Bq/kg]
^{137}Cs	12±9	1.1	55.0	18±9	0.6	45.6
^{238}U	51±9	24.0	72.0	45±16	18	90
^{226}Ra	39±7	19.7	51.0	33±9	17.1	53
^{232}Th	53±8	22.0	64.0	39±14	17.7	70
^{40}K	554±92	238	730	495±164	196	880

Conclusion

Contamination with long-lived produced and natural radionuclides is one of the factors which can disturb the historical ambient. It is very important to continue with the radioactivity measurements of soil in Vojvodina region in order to obtain the sufficient data for statistical processing.

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