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Determination of depleted uranium at the Novi Sad low-level laboratory

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Activity concentration of natural uranium has been determined in environmental samples at the low-level gamma-spectroscopy laboratory of the Faculty of Science in Novi Sad for over 20 years. When the issue of stating the presence of depleted uranium in such samples arose, the unique capability of our measuring equipment (GMX type of HPGe detector with enhanced efficiency below 100 keV, and iron low level shielding) was fully exploited. An appropriate technique for detecting depleted uranium was developed. This technique and the results of measurements of about 100 environmental samples (soil, plants, water, food) are presented and discussed.

KEY WORDS: *Uranium; Environmental Pollutants; Spectrometry, Gamma; Yugoslavia*

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INTRODUCTION

Low-level high-resolution gamma spectroscopy is a very convenient technique for determining the activity concentration of uranium in environmental samples in which the ^{238}U activity exceeds 1 Bq/kg. Our low-level gamma spectroscopy group in Novi Sad has performed such measurements for over two decades (1). The issue of the possible presence of depleted uranium in the environment has recently been of considerable public interest. We therefore developed a method for determining the presence of depleted uranium in a sample (2) and measured some 100 samples using this method.

EXPERIMENTAL TECHNIQUE

The radionuclide content of the samples was measured by means of the reversed electrode "GMX" - type HPGe spectrometer, manufactured by ORTEC. The nominal efficiency of the detector is 32% and the resolution is 1.9 keV. This detector has a thin dead layer on the outer surface and a beryllium entrance window, thus enabling the detection of gamma rays of energy below 100 keV, with excellent efficiency. The detector was calibrated using reference radioactive materials, in cylindrical geometry. Matrix effects were taken into account by means of a computer code.

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The detector was placed inside a 25 cm - thick iron shield made of pre - Second World War cast iron (3). Compared with lead, iron shields, due to the absence of the lead X-rays, offer a definite advantage in the low energy region. Samples were typically measured over a period of 50 ks.

DETERMINATION OF DEPLETED URANIUM

The natural abundance of uranium isotopes is presented in Table 1.

Table 1. Natural abundance of uranium isotopes

ISOTOPE	ABUNDANCE [%]	DECAY TYPE	HALF-LIFE [YEARS]
^{238}U	99.2745±0.0015	α	$(4.468±0.005)×10^9$
^{235}U	0.7200±0.0012	α	$(7.037±0.011)×10^8$
^{234}U	0.0055±0.0005	α	$(2.454±0.006)×10^5$

In principle, the lower content of ^{235}U in depleted uranium could be detected by gamma-spectroscopic determination of the $^{235}U/^{238}U$ ratio. However, at modest activity concentrations of uranium of about 100 Bq/kg, which is roughly twice that of the natural level in typical soil, direct measurements of the $^{235}U/^{238}U$ ratio cannot yield reliable information concerning the presence of depleted uranium in the sample (4).

On the other hand, use can be made of the fact that nuclear and chemical processing of natural uranium destroys the radioactive equilibrium between ^{238}U and ^{226}Ra (half-life 1500 y), as radium is removed from the material. Thus, the difference of the activity concentrations yields the activity concentration of depleted uranium:

A (^{238}U) - A (^{226}Ra) = A (DU)

In order to measure the difference between ^{238}U and ^{226}Ra, the activity of ^{238}U should be determined from the activity of ^{234}Th or ^{234}Pa, the first short-lived daughters of ^{238}U. The most prominent gamma rays of these nuclei are listed in Table 2.

Table 2. Principal gamma-ray lines of the first daughters of ^{238}U

NUCLEUS	E_γ [KEV]	I_γ [%]
^{234}Th	63.3	3.8
	92.3	2.7
	92.8	2.7
^{234}Pa	76.6	0.4
	98.4	0.2
	1001	0.9

It is clear from Table 2 that the 63.3 keV transition is the best analytical line for ^{238}U determination, provided the detector features good efficiency below 100 keV. As described earlier, our detection system is able to measure the intensity of this line very conveniently. The determination of ^{226}Ra is a much simpler task. The first daughter of ^{226}Ra is the radioactive noble gas ^{222}Rn which decays with a half life of 3.8 days to ^{214}Pb and ^{214}Bi exhibiting a considerable number of intensive analytical lines. Storage of samples in sealed holders for about 40 days restores the ^{222}Rn equilibrium and the activity of ^{214}Pb and ^{214}Bi becomes equal to that of ^{226}Ra.

The spectrum of a soil sample in which depleted uranium is present and the spectrum of a soil sample containing no depleted uranium are given in Figure 1, where the above-mentioned analytical lines are marked.

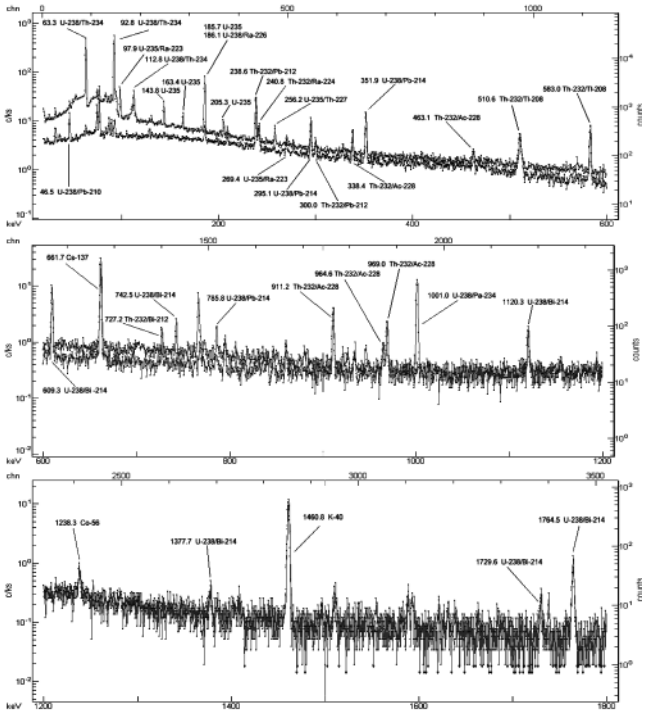

Figure 1. Spectra of soil samples with (upper spectrum) and without (lower spectrum) depleted uranium

The background that contains the same lines, especially post-radon lines, is not subtracted. The results of the application of the above-discussed method applied to three soil samples with a high, medium and zero depleted uranium content, are presented in Table 3.

Table 3. Depleted uranium in soil samples

Code	^{238}U [Bq/kg]	^{226}Ra [Bq/kg]	$^{238}U_d$ [Bq/kg]	^{40}K [Bq/kg]	^{232}Th [Bq/kg]
LVJ4	5000±130	19.8±2.1	4980±130	530±40	30±3
LVJ3	115±15	13.2±2.3	98±16	187±22	13.5±1.1
LVJ7	33±8	22.4±2.2	<10	1210±70	45±3

As can be noticed from the last row, for our detector, our background conditions and our measuring times of about 50 ks, the detection limit for depleted uranium is about 10 Bq/kg.

EXPERIMENTAL RESULTS AND CONCLUSIONS

The experimental results are presented in Tables 4 to 8.

Table 4. Activity concentration of characteristic radionuclides in soil samples in Novi Sad

Sample	Activity concentration [Bq/kg]					
	^{238}U	^{226}Ra	^{232}Th	^{40}K	U_d	^{137}Cs
Soil, TV Novi Sad I (06. '99)	<26	23.4±1.2	31±10	382±13	<26	<0.27
Soil, TV Novi Sad II (06. '99)	25±9	27.9±1.2	37.0±1.5	479±21	<9	0.6±0.4
Soil, oil refinery (reservoir) (04. '99)	11±6	10.6±0.4	11.0±0.5	219±11	<9	0.8±0.2
Soil, oil refinery (gate 4) (04. '99)	20±6	18.7±0.5	20.3±1.1	281±17	<6	1.0±0.4
Soil, oil refinery ("uljara 1") (05. '99)	20±8	16.1±.0.7	17.5±1.1	312±21	<6	0.5±0.3
Soil, oil refinery (workshop) (05. '99)	20±11	12.5±0.6	12.3±1.1	273±14	<11	3.2±0.6
Soil, Ribnjak (07. '99)	24±1	23.2±2.0	39.3±2.4	380±60	<2.2	<0.4
Soil, TV-tower, Venac (06. '99)	12±5	19.5±0.4	24.1±0.7	325±13	<5	0.6±0.1
Soil, EPS, Venac (09. '99)	27±20	30.3±0.9	41.8±2.2	434±23	<20	<0.4
Concrete, Žeželj bridge (04. '99)	14±5	10.4±0.3	10.9±0.8	151±10	<5	0.7±0.2
Sand, Naftagas (09.'99)	19±11	17±1	3±2	290±20	<11	0.8±0.4
Soil, Iriški venac 1a (09. '99)	80±30	46±2	62±4	560±50	<30	27±2
Soil, Iriški venac 1b (09. '99)	56±23	54±3	49±3	450±40	<23	86±5
Soil, Iriški venac 2 (09. '99)	80±30	46±2	62±4	580±40	<30	27±2
Soil, Iriški venac 7 (09. '99)	64±20	41±2	60±3	730±40	<20	29±2
Soil, Iriški venac 8 (09. '99)	80±30	46±2	62±4	580±40	<30	27±2
Soil, Iriški venac 10 (09. '99)	35±27	138±7	44±4	440±70	<28	188±13
Soil, Iriški venac 12 (09. '99)	54±17	39±2	55±4	680±40	<17	144±8
Sediment, canal DTD-mouth of Danube (09.'99)	33±10	31.3±1.4	30±2	459±27	<10	25.1±1.6
Sediment, Danube-Subić (09. '99.)	55±10	49±2	32±2	408±29	<10	46±2

Table 5. Activity concentration of characteristic radionuclides in water samples

Sample	Activity concentration [mBq/l]				
	^{238}U	^{226}Ra	^{232}Th	U_d	^{137}Cs
Water from crater (Ribnjak) (08. '99)	<6	<2	<26	<6	<2
Water from well vp1 (04. '99)	<7	<5	<4	<9	<1
Water from well vp2 (04. '99)	<8	<7	<5	<11	<2
Water from well vp3 (04. '99)	<8	<4	<3	<9	<1
Water from water supply (03. '99)	<4	<2	<2	<4	<1
Water from water supply (04. '99)	<4	<5	<2	<6	<2
Water from water supply (05. '99)	<4	<6	<2	<7	<1
Water from water supply (06. '99)	<4	<2	<3	<4	<1

Table 6. Activity concentration of characteristic radionuclides in plants and food

Sample	Activity concentration [Bq/kg]					
	^{238}U	^{226}Ra	^{232}Th	^{40}K	U_d	^{137}Cs
Green salad (17.05.1999)	<0.7	<0.7	<0.9	83±19	<1.0	<0.4
Green salad (09.06.1999)	<12	<0.6	<1.6	68±15	<12	0.5±0.4
Spinach (17.05.1999)	<0.7	<0.8	<1.0	231±22	<1.1	<0.4
Green onion (17.05.1999)	<1.5	<1.1	<1.4	57±21	<1.9	<0.6
Green onion (09.06.1999)	<16	<0.7	<1.0	27±13	<16	<0.4
Radish (17.05.1999)	<0.6	<0.6	<1.1	54±11	<0.8	<0.3
Radish (9.06.1999)	<13	<0.5	<0.8	41±13	<13	<0.3
Potatoes (9.06.1999)	<13	<0.7	<1.2	98±19	<13	<0.4
Pea (Ruski Krstur) (06. '99)	<14	<0.9	<1.2	28±20	<14	<0.5
Beans (Novi Sad) (06. '99)	<11	<0.5	<0.5	471±29	<11	0.4
Maize (07.06.1999)	<16	<0.6	<0.9	82±16	<16	0.4
Cherry (17.05.1999)	<0.6	<0.6	<0.7	52±13	<0.8	<0.4
Cherry (09.06.1999)	<0.5	<0.5	<0.9	44±12	<0.7	<0.4
Strawberry (17.05.1999)	<0.7	<0.4	<0.5	44±9	<0.8	<0.5
Strawberry (09.06.1999)	<10	<0.5	<0.7	22±19	<10	<0.6
Fresh milk (06.'99)	<0.5	<0.4	<1.0	69±18	<0.6	<0.4
Fresh white fish, Danube, Sr.Karlovci (21.05.1999)	<0.9	<0.8	<1.0	69±18	<1.2	<25
Fresh fish "babuska" Begečka jama (09. '99)	<1.6	4.7±0.2	<0.3	94±5	<1.6	0.3±0.1
Fish (pike, chub, sheath-fish) Begečka jama (09. '99)	<1.5	0.3±0.1	<0.2	108+4	<1.5	0.3±0.1
Fish (sterlet), Danube, Subić (09. '99)	<1.5	4.3±0.3	<0.3	70±4	<1.5	<0.3

Table 7. Activity concentration of characteristic radionuclides in wheat samples

Sample	Activity concentration [Bq/kg]					
	^{238}U	^{226}Ra	^{232}Th	^{40}K	U_d	^{137}Cs
Wheat 2s (30.08.1999)	<14	<1.5	<1.8	128±16	<14	<6
Wheat 15 Šid, "Mlintest" (30.08.1999)	<17	<1.2	1.0±0.9	150±19	<17	<4
Wheat 15s Šid, "Mlintest" (30.08.1999)	<18	<1.2	1.6±1.0	103±16	<18	<7
Wheat 22 Šid, "Mlintest" (30.08.1999)	<10	7.5±1.7	<1.3	138±17	<10	<6
Wheat 22s Irig, (30.08.1999)	<10	2.9±1.4	<1.5	128±16	<10	<5
Wheat 26s Kula, (30.08.1999)	<17	3.6±1.4	<1.6	140±17	<17	<5
Wheat 27s Ruski Krstur (30.08.1999)	<10	<1.4	<1.1	131±16	<10	<4
Wheat 30s Temerin, "D.D. Petefi" (30.08.1999)	<10	5.6±1.3	1.2±1.1	149±19	<10	<4
Wheat 42 Prigrevica, PIK (30.08.1999)	<8	<3.8	<1.3	149±16	<9	<4
Wheat 43 Kupusina, Z.Z. "Pčela" (30.08.1999)	<10	2.0±1.3	<1.4	121±18	<10	<5
Wheat 45 Z.Z. "Bezdan", (30.08.1999)	<12	<1.3	<0.9	132±16	<12	<4
Wheat 58 flour mill "Novi Kneževac", (30.08.1999)	<12	<1.7	<1.0	143±18	<12	<4
Wheat 160t (30.08.1999)	<10	1.4±1.2	<1.3	141±18	<10	<5

Table 8. Activity concentration of characteristic radionuclides in soil

Region	Sample	^{238}U [Bq/kg]	^{226}Ra [Bq/kg] post Rn	$^{238}U_d$ [Bq/kg]	^{40}K [kBq/kg]	^{232}Th [Bq/kg]
1	1	29 ± 7	15.7 ± 2.9	<18	0.450 ± 0.022	34.4±1.9
1	2	25 ± 6	15 ± 4	<15	0.480 ± 0.023	34.5±2.3
1	average 3 and 4	5000±1300	19.8 ± 2.1	5000±1300	0.53 ± 0.04	30±3
1	5	70 ± 12	29 ± 5	32±16	0.587± 0.028	46.5±2.5
1	6	42 ± 8	30 ± 4	<16	0.554±0.027	42.2±2.4
1	7	191±24	35 ± 4	146±27	0.73± 0.04	52.0±2.9
		201±25	52 ± 3	133±30	0.689± 0.297	51.2±2.7
1	8	32 ±11	24 ± 4	<15	0.522 ± 0.026	41.8±2.5
2	average	115±15	13.2± 2.3	98±16	0.187 ± 0.022	13.5±1.1
2	1	17 ± 10	8.3 ± 2.2	<17	0.128 ± 0.011	11.4±0.9
2	2	14 ± 5	9.6 ± 1.4	<8	0.116 ± 0.010	13.1±1.0
2	3	75± 10	10.8± 1.9	61±11	0.137 ± 0.010	12.4 ±0.9
2	4	19 ± 5	11.3 ± 1.8	<10	0.145 ± 0.010	14.0±1.0
2	5	16 ±6	7.5± 1.8	<13	0.141± 0.010	11.4±0.7
3	average	2000 ±600	116 ± 11	1850±600	1.28± 0.09	72 ± 7
3	1	133±19	82 ± 7	<70	0.76± 0.03	55 ± 3
		113 ± 18	76±6	<44	0.695± 0.028	52 ± 3
3	2	107± 14	67 ± 7	<57	0.81± 0.04	46.9 ± 2.6
3	3	115 ± 14	68 ± 6	<63	0.81 ± 0.03	49.7 ± 2.6
		108 ± 14	72±7	<53	0.76 ± 0.04	47.7 ± 2.6
3	4	56 ± 9	30 ± 4	17±14	0.510 ± 0.027	30.9 ± 2.0
4	average a)	42 ± 9	22 ± 3	13±12	1.14 ± 0.08	52 ± 3
4	average b)	74 ± 10	29.7± 2.8	35±14	1.15 ± 0.07	62 ± 4
4	1	38 ± 8	19 ± 3	<27	1.18 ± 0.08	48.6 ± 2.8
4	2	65 ± 16	26 ± 4	31±19	1.02 ± 0.07	67 ± 4
4	3	33 ± 8	22.4±2.2	<15	1.21 ± 0.07	45 ± 3
4	4	41 ± 8	26.4±2.6	<19	1.19 ± 0.08	51 ± 4
4	5	44 ± 7	24 ± 3	13±11	1.29 ± 0.08	31.7 ± 2.4
4	6	40 ± 11	20 ± 3	14±13	0.93 ± 0.06	41 ± 3
4	7	51 ± 9	25 ± 4	<36	0.85 ± 0.04	61 ± 3
4	8	47 ± 9	27.2±2.1	<24	0.98 ± 0.05	64 ± 4
1	4	20±8	18.4±1.6	<10	0.144±0.013	16.5±1.1
1	5	10±6	7.5±1.1	<7	0.170±0.017	8.9 ±0.7
1	2	21±6	12.7±1.9	<12	0.174±0.011	12.9±1.5
1	6	29±6	18±3	<15	0.186±0.011	18.5±1.2
2	1	24±8	16.1 ±1.9	<13	0.191±0.011	16.2±0.9
2	2	26±5	15.5±2.8	<14	0.209±0.011	16.0±0.9
2	3	18±8	12.9±2.2	<10	0.174±0.012	13.5±0.9
1	1	19±6	10.0±1.1	<13	0.159±0.011	9.8±0.7
1	4	20±8	18.4±1.6	<10	0.144±0.013	16.5±1.1
1	5	10±6	7.5±1.1	<7	0.170±0.017	8.9 ±0.7

As can be concluded from Tables 4 to 8, measurements of samples taken from the region of Novi Sad prove the absence of depleted uranium in them. Samples contaminated with depleted uranium are all from the southern part of Serbia.

Our method of detecting depleted uranium can be employed to further survey the radioactivity of the region and especially to study the migration of depleted uranium from the contaminated areas.

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