DEPLETED URANIUM DETERMINATION AT THE NOVI SAD LOW LEVEL FACILITY

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ABSTRACT

Natural uranium determination in environmental samples at the low-level gamma-spectroscopy laboratory of the Faculty of Science in Novi Sad has more than 20 years long tradition. When the issue of depleted uranium emerged the experimental advantages of the measuring equipment (GMX type of HPGe detector with enhanced efficiency below 100 keV, and iron low level shielding) where fully exploited. A detection technique selective for depleted uranium was developed. The details of this method together with the results for about 100 samples (soil, plants, water, food) are presented, and discussed.

Key words: depleted uranium, low-lewel gamma spectrometry, detection

INTRODUCTION

Low-level high-resolution gamma spectroscopy is a very convenient technique for uranium determination in environmental samples with ^{238}U activity concentration above 1 Bq/kg. The low-level gamma spectroscopy group in Novi Sad performed such measurements for more than two decades [IAEA89]. When the issue of depleted uranium determination emerged, responding to the strong public interest a method for depleted uranium determination was developed and applied for measurements on about 100 samples [Bikit98a].

THE EXPERIMENTAL TECHNIQUE

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

The detector was operated inside the 25 cm thick iron shield made from pre II world war casted iron [Bikit98]. As compared with led shields the iron shields have definite advantages in the low energy region due to the absence of the lead x-rays. The typical time of measurements of the samples was 50 ks.

DEPLETED URANIUM DETERMINATION

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

Table 1. Natural abundance of uranium isotopes

Isotope	Abundance [%]	Decay type	Half-life
^{238}U	99.2745±0.0015	α	(4.468±0.005)×10^{9} a
^{235}U	0.7200±0.0012	α	(7.037±0.011)×10^{8} a
^{34}U	0.0055±0.0005	α	(2.454±0.006)×10^{5} a

The lowered content of ^{235}U in depleted uranium can be in principle detected by gamma-spectroscopic determination of the ^{235}U/^{238}U ratio. But at modest activity concentrations of uranium of about 100 Bq/kg, which is roughly twice above the natural level in typical soil, the direct ^{235}U/^{238}U ratio measurement can not yield reliable information on the presence of depleted uranium in the sample [Manojlović90].
On the other hand one can make use of the fact that during the nuclear and chemical processing of natural uranium the radioactive equilibrium between ^{238}U and the 1500 y half-life ^{226}Ra is destroyed by the removal of radium from the material. Thus the difference of the activity concentrations yields the activity concentration of depleted uranium:

$$A (^{238}U) – A (^{226}Ra) = A (DU).$$

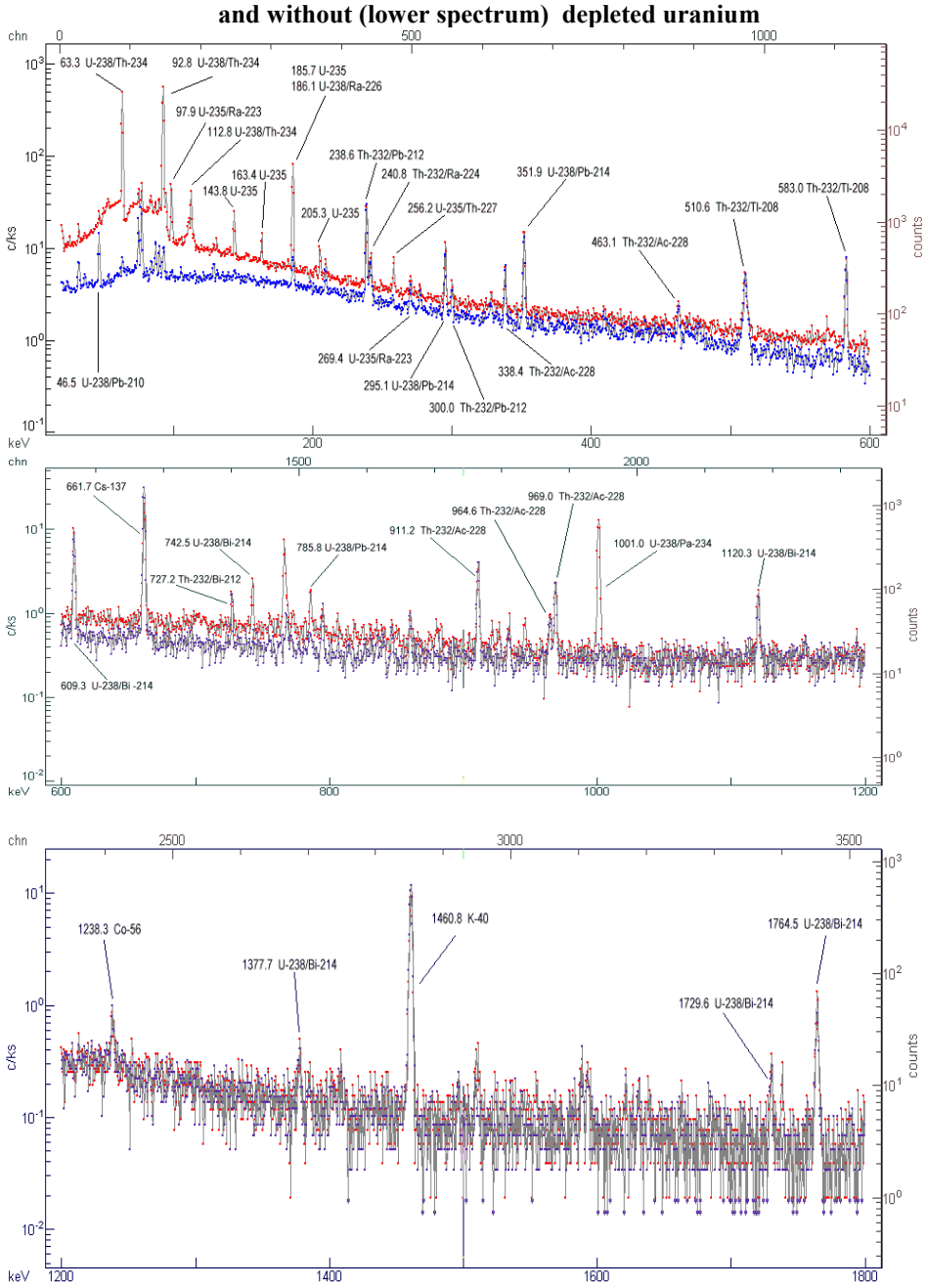
In order to measure the ^{238}U and ^{226}Ra difference the ^{238}U activity should be determined from the first short-lived daughters of ^{238}U, from the ^{234}Th or ^{234}Pa activity. The most prominent gamma rays of these nuclei are presented on Table 2.

Table 2. The most prominent gamma-rays of first ^{238}U daughters

Nuclei	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

As can be noticed from the Table 2. for detectors with good efficiency below 100 keV, the 63.3 keV transition is the best analytical line for ^{238}U determination. Our detection system described earlier is very convenient for the measurement of the intensity of this line. The

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



determination of ^{226}Ra is a much simpler task. The first daughter of ^{226}Ra is a noble radioactive gas ^{222}Rn which decays with the half life of 3.8 days to ^{214}Pb and ^{214}Bi having lot of intensive analytical lines. When keeping the samples in sealed sample holders for about 40 days the ^{222}Rn equilibrium will be restored and the ^{214}Pb and ^{214}Bi activity will be equal to the ^{226}Ra activity. A soil sample contaminated with depleted uranium compared with the soil sample without depleted uranium is presented on Figure 1.
The background, which contains the same lines especially post radon lines, is not subtracted. The results of the application of the method described for three soil samples with high, medium and zero depleted uranium content are presented in Table 3.

Table 3. Depleted uranium in soil samples

Sample	^{238}U [Bq/kg]	^{226}Ra [Bq/kg]	$^{238}U_d$ [Bq/kg]	^{40}K [kBq/kg]	^{232}Th[Bq/kg]
LVJ4	$(5.0\pm1.3)\cdot10^3$	19.8±2.1	$(5.0\pm1.3)\cdot10^3$	0.53±0.04	30±3
LVJ3	115±15	13.2±2.3	98±16	0.187±0.022	13.5±1.1
LVJ7	33±8	22.4±2.2	<10	1.21±0.07	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. -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 (o6. '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 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
Maise (07.06. '99.)	<16	<0.6	<0.9	82±16	<16	0.4
Cherry (17.05. '99.)	<0.6	<0.6	<0.7	52±13	<0.8	<0.4
Cherry (09.06. '99.)	<0.5	<0.5	<0.9	44±12	<0.7	<0.4
Strawberry (17.05.'99.)	<0.7	<0.4	<0.5	44±9	<0.8	<0.5
Strawberry (09.06. '99.)	<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.'99.)	<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 6. 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) (08.1999.)	<6	<2	<26	<6	<2
Water from well vp1 (04.1999.)	<7	<5	<4	<9	<1
Water from well vp2 (04.1999.)	<8	<7	<5	<11	<2
Water from well vp3 (04.1999.)	<8	<4	<3	<9	<1
Water from water supply (03. 1999.)	<4	<2	<2	<4	<1
Water from water supply (04. 1999.)	<4	<5	<2	<6	<2
Water from water supply (05. 1999.)	<4	<6	<2	<7	<1
Water from water supply (06. 1999.)	<4	<2	<3	<4	<1

Table 7. Activity concentration of characteristic radionuclides in wheat samples

Sample	Activity concentration [Bq/kg]					
	^{238U}	^{226}Ra	^{232T}h	^{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 Mlin "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 samples from southern Serbia

REGION	*Samples*	^{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 i 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 ±24 201 ± 25	35 ± 4 52 ± 3	146±27 133±30	0.73 ± 0.04 0.689 ± 0.297	52.0 ± 2.9 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
REGION	***Samples***	**^{238}U [Bq/kg]**	**^{226}Ra [Bq/kg] post Rn**	**$^{238}U_d$ [Bq/kg]**	**^{40}K [kBq/kg]**	**^{232}Th [Bq/kg]**
3	average	2000 ±600	116 ± 11	1850±600	1.28 ± 0.09	72 ± 7
3	1	133±19 113 ± 18	82 ± 7 76±6	<70 <44	0.76 ± 0.03 0.695 ± 0.028	55 ± 3 52 ± 3
3	2	107 ± 14	67 ± 7	<57	0.81 ± 0.04	46.9 ± 2.6
3	3	115 ± 14 108 ± 14	68 ± 6 72±7	<63 <53	0.81 ± 0.03 0.76 ± 0.04	49.7 ± 2.6 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
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	3	19±5	15.1±2.0	<11	0.158±0.015	17.4±1.0
2	4	24±6	17.3±2.5	<10	0.162±0.010	15.5±1.1

REGION	***Samples***	**^{238}U [Bq/kg]**	**^{226}Ra [Bq/kg] post Rn**	**$^{238}U_d$ [Bq/kg]**	**^{40}K [kBq/kg]**	**^{232}Th [Bq/kg]**
-	5A	4000±700	142±10	3800±700	0.90±0.04	72±7
-	5B	1700±400	138±8	*1500±400*	0.94±0.04	75±4
-	6	43±9	46.8±2.1	<17	0.43±0.017	25.9±1.3
-	4	70±9	57.5±2.2	<20	0.53±0.03	35.7±1.9
-	3	120±18	103±5	<45	0.83±0.04	46.3±2.3
-	2	81±10	58.8±2.4	<25	0.78±0.04	34.5±1.8
-	1	137±21	112±5	<49	0.91±0.04	61±4

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

Our method for depleted uranium detection can be employed for further surveying the radioactivity of the region and especially for the investigation of depleted uranium migration from the contaminated areas.

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