

Environmental radioactivity in southern Serbia at locations where depleted uranium was used

Nataša B. Sarap, Marija M. Janković, Dragana J. Todorović, Jelena D. Nikolić, and
Milojko S. Kovačević

*University of Belgrade, Vinča Institute of Nuclear Sciences, Radiation and Environmental Protection Department,
Belgrade, Serbia*

Received in July 2013
CrossChecked in February 2014
Accepted in April 2014

In the 1999 bombing of the Federal Republic of Yugoslavia, NATO forces used ammunition containing depleted uranium. The cleaning of depleted uranium that followed was performed in southern Serbia by the Vinča Institute of Nuclear Sciences between 2002 and 2007 at the locations of Pljačkovica, Borovac, Bratosele, and Reljan. This paper presents detailed results of radioactivity monitoring four years after cleaning (2011), which included the determination of gamma emitters in soil, water, and plant samples, as well as gross alpha and beta activities in water samples. The gamma spectrometry results showed the presence of natural radionuclides ^{226}Ra , ^{232}Th , ^{40}K , ^{235}U , ^{238}U , and the produced radionuclide ^{137}Cs (from the Chernobyl accident). In order to evaluate the radiological hazard from soil, the radium equivalent activity, the gamma dose rate, the external hazard index, and the annual effective dose were calculated. Considering that a significant number of people inhabit the studied locations, the periodical monitoring of radionuclide content is vital.

KEY WORDS: *environmental samples; gamma spectrometry; gross alpha activity; gross beta activity*

Over the last 40 years, Europe has experienced several major radioactive fallouts, from atmospheric nuclear tests in North Africa during the 1960s, the Chernobyl accident in 1986, the Acerinox accident in 1998 to the wars in former Yugoslavia in the 1990s (1). Depleted uranium (DU) came into military use in the early 1990s and was first used in 1991 in the Gulf War (2, 3). The DU used during the wars in former Yugoslavia represents the most recent source of radioactivity released into the environment, following NATO air strikes (1, 4-6). NATO and Serbian Military (VS) data on DU use are in large discordance, but overlap for at least four locations (5, 6) in southern Serbia: Pljačkovica, Borovac, Bratosele, and Reljan (Figure 1).

Uranium is a naturally occurring ubiquitous heavy metal found in various chemical forms in all

soils, rocks, seas, and oceans. It is also present in drinking water and food. Natural uranium consists of a mixture of three different isotopes: ^{238}U (99.27 %), ^{235}U (0.72 %), and ^{234}U (0.0054 %) (7). It is both chemically toxic and radioactive, and being water-soluble, it is easily taken up by plants, thus entering food chains and soil/water systems (8). Metallic uranium used for military purposes is practically insoluble in water and body fluids. However, while exposed to air and water, uranium is slowly oxidised to a +4 or +6 state (9).

The uranyl ion easily forms complexes, mainly carbonate $[\text{UO}_2(\text{CO}_3)_2]^{-2}$, which are well dissolved in water and body fluids. As the result of these chemical processes (weathering), uranium dispersed into the environment in a metallic form can slowly migrate to drinking water and plants and enter the

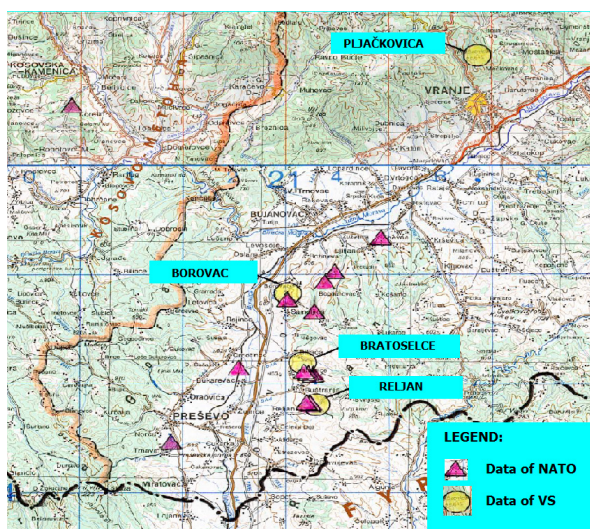


Figure 1 Information on locations in southern Serbia which are targeted depleted uranium. GPS coordinates of locations: Pljačkovica (42° 34' 47" N; 21° 53' 50" E); Borovac (42° 23' 45" N; 21° 45' 09" E); Bratosele (42° 20' 40" N; 21° 45' 23" E); Reljan (42° 18' 53" N; 21° 45' 58" E)

human food chain. However, the weathering of DU penetrators strongly depends on the type of soil, its humidity and temperature (9). Depending on the physicochemical characteristics of the medium and chemical form in which uranium is bound, there are various scenarios for environmental contamination.

Quantitative determination of the DU fraction in environmental samples is difficult, primarily because of the relatively large content of natural uranium present in the environment. Based on the obtained concentrations of ^{235}U and ^{238}U and their ratio, a conclusion whether this ratio presents the natural activity ratio of uranium isotopes or possible presence of DU can be reached.

Considering the fact that people live and work on the locations examined here, it is necessary to periodically monitor the content of radionuclides in samples from these locations, especially because of the use of DU. Therefore, one of the aims of this study was to offer a detailed approach to perform such monitoring.

MATERIALS AND METHODS

Environmental monitoring

Regular radioactivity monitoring in Serbia includes spectrometric measurements of gamma emitters in soil, water and plant samples, as well as determination of the gross alpha and beta activities

in water samples. The cleaning of DU left over after NATO air strikes, along with detailed dosimetric screening and decontamination, was performed by the Radiation and Environmental Protection Department of the Vinča Institute of Nuclear Sciences from 2002 to 2007 at Pljačkovica, Borovac, Bratosele, and Reljan, located in southern Serbia, in coordination with the Agency for Radiation Protection and Nuclear Safety of Serbia (10) (Figure 1). Because of the loose soil at these locations, projectiles containing DU were found already at a depth of one meter. The discovered projectiles, contaminated soil, and all radioactive materials were disposed of as radioactive waste. After the cleaning, the terrain was aligned and another dosimetric screening was performed. In 2011, the Department collected and analysed samples from these locations in order to determine the activity concentrations of radionuclides and verify that the fields were successfully cleaned of DU.

Sampling and sample preparation

Soil samples were collected at 5 measuring points for each location (east, west, north, south, and centre) at a depth of 10-15 cm using a probe. The sampling depth was chosen based on the fact that these measurements follow the migration of radionuclides by depth (11). About 2 kg of samples were placed into polyethylene bags and transported to the laboratory. After removing the stones and vegetation, all soil samples were dried to 105 °C, sieved, placed in plastic 500 mL Marinelli beakers and left to reach radioactive equilibrium for four weeks (12).

The plant samples were taken differently and sampling included the presence of plant species (hay and grass). About 1 kg of samples was placed into polyethylene bags and transported to the laboratory. Plant samples were dried at room temperature over a few days, ashed at 450 °C during 24 h, placed in plastic boxes of 100 mL and left to reach radioactive equilibrium for four weeks (12).

Water samples were taken from public fountains and wells from where the inhabitants draw water. These samples (20 L each) were collected directly from the springs in plastic bottles and tightly closed. For gamma spectrometric measurements, a volume of 17 L of water samples was acidified to pH 2, evaporated to 200 mL under an infrared lamp, and then poured into 200 mL cylindrical polyethylene vials. The samples were then stored to reach radioactive equilibrium (12). For measurements of

gross alpha and beta activity, a volume of 3 L of water samples was evaporated to dry residue under an infrared lamp. The remainder was heated to dryness at 450 °C (13, 14). The residues were transferred quantitatively to a stainless-steel planchet and measurements were performed immediately after preparation.

Plant samples were concentrated by mineralisation and water samples by evaporation in order to maximize the detection efficiency, which is higher in geometries that are not collected in a large volume because the self-absorption effect is smaller.

Procedure for gamma spectrometry

Gamma spectrometric measurements were performed using HPGe Canberra detectors (Canberra Industries, Meriden, Connecticut, USA) with counting efficiency of 18 and 20 %. Due to the low activity in the samples and to reduce the uncertainty of measurement, the samples were measured 60,000 s. The calibration of detectors for water and plant sample measurement was performed using a cylindrical polyethylene bottle of 200 mL and a plastic box of 100 mL in volume, respectively. This secondary reference material was obtained from the primary reference liquid radioactive material (9031-OL-116/08, type ERX, Czech Metrological Institute, Prague, Czech Republic) spiked with a series of radionuclides (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{88}Y , ^{113}Sn , ^{85}Sr , ^{137}Cs , and ^{210}Pb) with total activity of 114.9 kBq on 3 March 2008. The calibration of detectors for measurement of soil samples was performed using a silicone resin matrix in the geometry of a plastic Marinelli beaker of 500 mL in volume, (9031-OL-208/08, type ERX, Czech Metrological Institute, Prague, Czech Republic) spiked with a series of radionuclides (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{203}Hg , ^{88}Y , ^{113}Sn , ^{85}Sr , and ^{137}Cs) with total activity of 40.624 kBq on 15 April 2008.

The spectra were analysed using the program GENIE 2000 (Canberra Industries, Meriden, Connecticut, USA). The activity of ^{226}Ra and ^{232}Th was determined by their decay products: ^{214}Bi (609 keV, 1120 keV, and 1764 keV), ^{214}Pb (295 keV and 352 keV) and ^{228}Ac (338 keV and 911 keV), respectively. ^{235}U was determined via 186 keV corrected for ^{226}Ra . ^{238}U was determined via ^{234}Th (63 keV) or by ^{234}Pa ($t_{1/2}=1.17$ min, 1000 keV). The activities of ^{40}K and ^{137}Cs were determined from its 1460 keV and 661 keV, respectively. The activities of ^{210}Pb and ^7Be were determined from its 46 keV and

477 keV, respectively. The background spectrum was recorded regularly after or before the sample counting, with an empty 200 mL cylindrical polyethylene bottle, 100 mL plastic box, and 500 mL plastic Marinelli beaker.

The accuracy and reproducibility of gamma spectrometry systems were verified on weekly basis by a quality control (QC) procedure. QC procedure consisted of the following steps: total background count rate was monitored to verify that the detector and shield have not been contaminated by radioactive materials; the total activity of the calibration source [^{60}Co and ^{133}Ba issued by the Czech Metrological Institute and traceable to BIPM (*Bureau International des Poids et Mesures*)] was used to check the efficiency calibration and the general operating parameters of the gamma spectrometry system (source positioning, contamination, library values, and energy calibration); the detector-shield background, full energy peak efficiency, peak shape, and peak drift were measured to confirm whether they were within the warning and acceptance limits; the acceptance limits were set according to the manufacturer's specifications (peak shape and drift, full energy peak efficiency) and previous experiences (detector shield background); the specific activity of the radionuclides in the samples (A) was calculated using the equation:

$$A = \frac{N}{t \times P_\gamma \times ef \times V(m)} \quad (1)$$

where N is count rate of the sample, t – counting time (s), P_γ – probability of gamma decay (%), ef – detector efficiency (%), V and m are the volume (L) and mass (kg) of the sample.

Minimum detectable activity (MDA) was calculated by the equation (2):

$$MDA = \frac{LLD}{t \times P_\gamma \times ef \times V(m)} \quad (2)$$

where LLD is the detection limit, $LLD = 2.71 + 4.65\sqrt{B}$ and B is background.

The combined measurement uncertainty of results was calculated at the 95 % level of confidence ($k=2$).

Minimum detectable activity was inversely proportional to the measurement time of samples (equation 2), which is very important for spectrometry of gamma emitters in environmental samples. Also, in order to reduce the measurement uncertainty of the count rate below the photo peak of a certain energy that corresponds to radionuclides with low activities (such as ^{235}U), it is necessary to increase the

measurement time. Uncertainty measurement of count below the photo peak affects the overall uncertainty of the results, which can be calculated by the equation (3):

$$\sigma = \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta P_\gamma}{P_\gamma}\right)^2 + \left(\frac{\Delta ef}{ef}\right)^2 + \left(\frac{\Delta V(\Delta m)}{V(m)}\right)^2 + \sigma_n^2}$$

where Δ/N is relative uncertainty of count which is corrected on background, $\Delta t/t$ relative uncertainty of time measurement, $\Delta P_\gamma/P_\gamma$ relative uncertainty of the gamma decay probability (table value), $\Delta ef/ef$ relative uncertainty of detector efficiency, $\Delta V/V$ ($\Delta m/m$) relative uncertainty of volume determination (or mass determination), σ_n uncertainty due to the fitting and due to the unfairness in the calibration.

Procedure for gross alpha and gross beta activity

Gross alpha and beta activity in water samples were determined by α/β low-level proportional counter Thermo Eberline FHT 770 T (ESM Eberline Instruments GmbH, Erlangen, Germany). The counting time was 7,200 s.

Calibration was performed by using a standard source of ^{90}Sr (EM 145, Prague, Czech Republic) with an activity of 189.4 Bq on 1 August 2011 for beta activity and a standard source of ^{241}Am (EM 445, Prague, Czech Republic) with an activity of 224 Bq on the 1 August 2011 for alpha activity. The counting gas was a mixture of 90 % argon and 10 % methane. The counting efficiencies for the system were 26 % for alpha and 35 % for beta. The background of each detector was determined by counting an empty planchet for 3,600 s.

Quality of sampling and measurement and the calculated uncertainty are very important for predicting the dose for a population. The accuracy and reproducibility of a gas proportional counter were verified on a periodic basis (every week). Calibration was done every week with calibration standards and efficiency was checked. Total background count rate without a source was monitored to verify that the detector and shield have not been contaminated by radioactive materials. Alpha and beta efficiencies of gas proportional counter were checked with ^{241}Am and ^{90}Sr sources, respectively.

Gross alpha and beta activity was calculated using the following formula:

$$A_{\alpha,\beta} = \frac{I}{V} \quad (4)$$

where $A_{\alpha,\beta}$ is the activity of the sample (Bq L^{-1}), V is the volume of the sample (L) that corresponds to the mass of solid residue, and I is given by the formula:

$$I = \frac{(N - B)}{ef} \quad (5)$$

where N is the count rate for the sample (s^{-1}), B is background (s^{-1}) and ef is the efficiency of the detectors for alpha and beta measurements.

Minimum detectable activity was calculated by the equation (6):

$$MDA = \frac{LLD}{V} \quad (6)$$

where LLD is the detection limit (s^{-1}) and V is the volume of the sample (L). Measurement uncertainty determined as expanded measurement uncertainty was 30–40 % for alpha and 15 % for beta.

Radium equivalent activity

The distribution of ^{226}Ra , ^{232}Th , and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra , ^{232}Th , and ^{40}K (8). It is calculated using the following equation (15, 16):

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K \quad (7)$$

where A_{Ra} , A_{Th} , and A_K are the specific activities of ^{226}Ra , ^{232}Th , and ^{40}K in Bq kg^{-1} , respectively.

Gamma dose rate calculation

The external gamma dose rate in the air 1 m above ground level was calculated from the measured specific activities of ^{226}Ra , ^{232}Th , and ^{40}K in soil assuming that the other radionuclides, such as ^{137}Cs , ^{90}Sr , and the ^{235}U series, can be neglected as they contribute very little to the total dose from environmental background (8, 17). The calculations were performed according to the following equation (18):

$$\dot{D} = 0.462 \times A_{Ra} + 0.604 \times A_{Th} + 0.042 \times A_K \quad (8)$$

where \dot{D} is the dose rate in nGy h^{-1} .

Calculation of external hazard index

The external hazard index, H_{ex} , was defined as (15):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (9)$$

The value of this index must be less than unity in order to keep the radiation hazard insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity (370 Bq kg⁻¹) (8).

Calculation of annual effective dose

To estimate the annual effective dose, the following must be taken into account: the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor (8). Using the dose rate data obtained from the concentration values of natural radionuclides in soil, adopting the conversion factor of 0.7 Sv Gy⁻¹ (18) from the absorbed dose rate in air to the effective dose received by adults, and considering that people in Serbia on average spend 20 % of their time outdoors, the annual effective doses were calculated by (8):

$$D_E = 0.7 \times 365 \times 24 \times 0.2 \times \dot{D} \quad (10)$$

where D_E is the annual effective dose in nSv.

RESULTS AND DISCUSSION

The results of measurements of gamma emitters (mean activity concentrations) in soil samples collected at different locations in southern Serbia are presented in Table 1 along with a comparison with relevant literature. The activity concentrations of the radionuclides ranged between 16-102 Bq kg⁻¹ for ²²⁶Ra, 21-90 Bq kg⁻¹ for ²³²Th, 184-979 Bq kg⁻¹ for ⁴⁰K, 2.9-19 Bq kg⁻¹ for ¹³⁷Cs, 1.1-7.4 Bq kg⁻¹ for ²³⁵U, and 21-95 Bq kg⁻¹ for ²³⁸U.

The man-made radioisotope ¹³⁷Cs, which has a generally negative impact on the environment, was identified in all of the samples and these values were in good agreement with those obtained in other regional studies (8, 19, 20). From the late 1980s, most ¹³⁷Cs originates from the Chernobyl nuclear accident.

The highest concentrations of ²²⁶Ra, ⁴⁰K, ¹³⁷Cs, ²³⁵U, and ²³⁸U were measured for Bratoselce, and the lowest for Borovac. Based on the measured concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K, radium equivalent activity, gamma dose rate, external hazard index and annual effective dose are calculated and presented in Table

2. The radium equivalent activity values for the soil samples varied from 60 to 260 Bq kg⁻¹. These values were below the recommended level (370 Bq kg⁻¹) (21). The mean gamma dose rate in air was calculated to be 78 nGy h⁻¹ and was insignificantly higher than the global average (57 nGy h⁻¹) (18). The calculated values of outdoor annual effective dose ranged from 0.03 to 0.15 mSv, which is close to the global average value (0.07 mSv) (18). The external hazard index obtained in this study ranged from 0.16 to 0.70. Since these values were lower than unity (18), we can conclude that the radiation hazard in the studied area was low.

The results of gamma emitter measurements in water samples from southern Serbia are presented in Table 3. ²²⁶Ra was detected in all of the water samples, except for Bratoselce. ⁴⁰K was detected in water samples from Pljačkovica and Reljan. The obtained values were low and mainly typical for water samples, while the activity concentrations for other radionuclides were below the detection limit.

The results on gross alpha and beta activity in the same water samples are presented in Table 4. Gross alpha activity ranged between <MDA–0.024 Bq L⁻¹, while gross beta activity ranged from 0.07 to 0.36 Bq L⁻¹. The obtained results showed that the natural activity concentrations of alpha and beta emitting radionuclides in water samples from southern Serbia were within recommended levels (0.5 and 1.0 Bq L⁻¹, respectively) (22). The gross alpha activity (Table 4) was lower than the ²²⁶Ra concentration in the tested samples (Table 3), as the determination of ²²⁶Ra via daughters in cylindrical polyethylene bottle often leads to erroneous results. Gaseous Rn escapes from the water to a gaseous phase and remains partially present in the bottle, while its solid progenies ²¹⁴Bi and ²¹⁴Pb can non-uniformly deposit on the walls of a bottle.

The results of gamma emitters in plant samples are presented in Table 5. In addition to the radionuclides already mentioned, the plant sample also revealed a presence of ²¹⁰Pb and ⁷Be. The obtained results for ²¹⁰Pb are characteristic for plant samples and higher than values for other natural radionuclides. This was expected because a plant can absorb ²¹⁰Pb from soil and air. The activity concentrations of the radionuclides ranged between 1.8-4.5 Bq kg⁻¹ for ²²⁶Ra, 2.1-3.2 Bq kg⁻¹ for ²³²Th, 284-434 Bq kg⁻¹ for ⁴⁰K, <MDA-1.3 Bq kg⁻¹ for ¹³⁷Cs, 63-123 Bq kg⁻¹ for ⁷Be, and 7-50 Bq kg⁻¹ for ²¹⁰Pb. The obtained activity concentrations for ²³⁵U and ²³⁸U in plant samples from

Table 1 Activity concentrations of radionuclides in soil samples in southern Serbia and comparison with literature ($Bq\ kg^{-1}$)

Location	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	^{235}U	^{238}U	$^{235}U/^{238}U$
Pljačkovica	27±5	32±4	498±44	14±1	2.2±0.3	33±7	0.067
Borovac	16±3	21±3	184±19	3±1	1.1±0.2	21±5	0.052
Bratoselece	102±28	58±6	979±133	19±2	7.4±0.8	95±18	0.059
Reljan	35±6	90±11	941±64	5±1	2.9±0.4	49±12	0.059
Average	45	50	651	10	3.4	57	/
Comparison with literature							
Republic of Srpska (8)	47	41	536	26	3.4	64	/
Croatia (19)	74	62	650	39	5.4	110	/
Slovenia (19)	73.8	/	318	25.1	/	69.5	/
Vojvodina (Serbia) (20)	40	53	554	12	/	51	/
World (18)	35	30	400	/	/	35	/

Table 2 Radium equivalent activity, gamma dose rate, annual effective dose, and external hazard index at four locations in southern Serbia

Location	Radium equivalent activity ($Bq\ kg^{-1}$)	Gamma dose rate ($nGy\ h^{-1}$)	Annual effective dose (mSv)	External hazard index
Pljačkovica	111	53	0.07	0.30
Borovac	60	27	0.03	0.16
Bratoselece	260	123	0.15	0.70
Reljan	236	110	0.13	0.64
Average	167	78	0.10	0.45

Table 3 Activity concentrations of radionuclides in water samples from southern Serbia ($Bq\ L^{-1}$)

Location	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	^{235}U	^{238}U
Pljačkovica	Bazovnik	<0.02	<0.02	<0.13	<0.005	<0.007
	draw well	0.055±0.007	<0.02	0.17±0.03	<0.005	<0.007
Borovac	house	<0.02	<0.02	<0.13	<0.005	<0.007
	fountain	0.033±0.005	<0.02	<0.13	<0.005	<0.007
Bratoselece	courtyard 1	<0.02	<0.03	<0.13	<0.004	<0.009
	courtyard 2	<0.02	<0.02	<0.13	<0.005	<0.007
Reljan	fountain	0.018±0.003	<0.02	<0.13	<0.004	<0.009
	courtyard	0.040±0.007	<0.02	0.15±0.03	<0.005	<0.007

Table 4 Gross alpha and gross beta activity concentrations of water samples from southern Serbia ($Bq\ L^{-1}$)

Location	Gross alpha activity	Gross beta activity
Pljačkovica	Bazovnik	0.009±0.001
	draw well	0.020±0.003
Borovac	house	<0.006
	fountain	0.009±0.001
Bratoselece	courtyard 1	<0.006
	courtyard 2	<0.006
Reljan	fountain	<0.006
	courtyard	0.024±0.004

Table 5 Activity concentrations of gamma emitters in plant samples from southern Serbia (Bq per kg of dry matter)

Radionuclide	Location			
	Pljačkovica	Borovac	Bratoselce	Reljan
²²⁶ Ra	1.8±0.2	2.3±0.5	4.5±0.7	2.0±0.4
²³² Th	3.0±0.6	3.2±0.6	2.1±0.5	3.2±0.8
⁴⁰ K	434±43	308±32	284±31	431±45
¹³⁷ Cs	1.0±0.1	1.3±0.3	<0.4	0.8±0.2
²³⁵ U	<0.3	0.3±0.1	<0.2	0.3±0.1
²³⁸ U	<4	4.4±1.5	<2.2	3.5±1.4
⁷ Be	123±12	116±15	108±10	63±7
²¹⁰ Pb	50±6	32±4	33±4	7±2

Three plant samples were taken from each location

Pljačkovica and Bratoselce were below the detection limit.

CONCLUSION

The results of gamma spectrometric measurements of soil samples showed that the radioactivity levels in samples were similar to values characteristic for other locations in world where depleted uranium had not been used (8, 19, 20, 23–29).

According to the calculated values of external hazard index, there is no risk for the population living in the investigated area. Measurements of radioactivity in environmental samples at these locations in southern Serbia should, however, continue.

Acknowledgement

This work was partially supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, under Project no. III 43009.

REFERENCES

- Esposito M, Polić P, Bartolomei P, Benzi V, Martellini M, Cvetković O, Damjanov V, Simić M, Žunić Z, Živančević B, Simić S, Jovanović V. Survey of natural and anthropogenic radioactivity in environmental samples from Yugoslavia. *J Environ Radioact* 2002;61:271-82. PMID: 14689992
- Hamilton EI. Depleted uranium (DU): a holistic consideration of DU and related matters. *Sci Total Environ* 2001;281:5-21. PMID: 11778960
- Bleise A, Danesi PR, Burkart W. Properties, use and health effects of depleted uranium (DU): a general overview. *J Environ Radioact* 2003;64:93-112. doi: 10.1016/S0265-931X(02)00041-3
- McLaughlin JP, Vintro LL, Smith KJ, Mitchell PI, Žunić ZS. Actinide analysis of a depleted uranium penetrator from a 1999 target site in southern Serbia. *J Environ Radioact* 2003;64:155-65. PMID: 12500802
- Radenković MB, Kandić AB, Vukanac IS, Joksić JD, Đorđević DS. Chemical and radiochemical characterization of depleted uranium in contaminated soils. *Russ J Phys Chem A* 2007;81:1448-51. doi: 10.1134/S0036024407090191
- Radenković MB, Cupać SA, Joksić JD, Todorović DJ. Depleted uranium mobility and fractionation in contaminated soil (Southern Serbia). *Env Sci Pollut Res* 2008;15:61-7. PMID: 18306889
- Betti M. Civil use of depleted uranium. *J Environ Radioact* 2003;64:113-9. PMID: 12500798
- Janković M, Todorović D, Savanović M. Radioactivity measurements in soil samples collected in the Republic of Srpska. *Radiat Meas* 2008;43:1448-52. doi: 10.1016/j.radmeas.2008.03.004
- Bem H, Bou-Rabee F. Environmental and health consequences of depleted uranium use in the 1991 Gulf War. *Environ Int* 2004;30:123-34. PMID: 14664872
- Agency for Radiation Protection and Nuclear Safety of Serbia. Rulebook: On radioactivity monitoring. *Official Gazette of the Republic of Serbia*: 97/2011; 2011.
- Todorović D, Kovačević M. Study of investigation of radionuclides content in environmental at locations where depleted uranium was used. *Vinča Institute of Nuclear Sciences: Belgrade, Serbia*; 2011.
- HASL-300. The EML Procedures Manual. New York: Environmental Measurements Laboratory; 1983.
- International Atomic Energy Agency (IAEA). Measurements of Radionuclides in Food and the Environment. Technical Report Ser. No. 295. Vienna: IAEA; 1989.
- United States Environmental Protection Agency (US EPA). Prescribed Procedures for Measurement of Radioactivity in Drinking Water. EPA-600/4-80-032, Method 900.0.; 1980 [displayed 27 March 2014]. Available at <http://www.health.state.mn.us/divs/phl/accreditation/docs/refeparadioactivitydw.pdf>
- Beretka J, Mathew PJ. Natural radioactivity of Australian building materials, industrial wastes and by-products. *Health Phys* 1985;48:87-95. doi: 10.1097/00004032-198501000-00007
- Yu KN, Guan ZJ, Stokes MJ, Young ECM. The assessment of the natural radiation dose committed to the Hong Kong people. *J Environ Radioact* 1992;17:31-48. doi: 10.1016/0265-931X(92)90033-P

17. Kocher DC, Sjoreen AL. Dose-rate conversion factors for external exposure to photon emitters in soil. *Health Phys* 1985;48:193-205. PMID: 3972585
18. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources and effects of ionising radiation. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, 2008 [displayed 27 March 2014]. Available at http://www.unscear.org/docs/reports/2008/09-86753_Report_2008_Annex_B.pdf
19. Vaupotič J, Barišić D, Kobal I, Lulić S. Radioactivity and radon potential of the terra rossa soil. *Radiat Meas* 2007;42:290-7. doi: 10.1016/j.radmeas.2007.01.034
20. Bikit I, Slivka J, Čonkić Lj, Krmar M, Vesković M, Žikić-Todorović N, Varga E, Ćurčić S, Mrđa D. Radioactivity of the soil in Vojvodina (northern province of Serbia and Montenegro). *J Environ Radioact* 2005;78:11-9. doi: 10.1016/j.jenvrad.2004.03.034
21. Organization for Economic Cooperation and Development (OECD). Exposure to radiation from the natural radioactivity in building materials. Report by a Group Experts of the OECD Nuclear Energy Agency, 1979 [displayed 27 March 2014]. Available at <https://www.oecd-neo.org/rp/reports/1979/exposure-to-radiation-1979.pdf>.
22. Official Gazette of the Republic of Serbia Rulebook: On limits of radionuclides content in drinking water, foodstuffs, feeding stuffs, medicines, general use products, construction materials and other goods that are put on market. 86/2011; 2011.
23. Saleh H, Abu Shayeb M. Natural radioactivity distribution of southern part of Jordan (Ma'an) Soil. *Ann Nucl Energy* 2014;65:184-9.
24. Lu X, Zhao C, Chen C, Liu W. Radioactivity level of soil around Baqiao coal-fired power plant in China. *Radiat Phys Chem* 2012;81:1827-32. doi: 10.1016/j.radphyschem.2012.07.013
25. Montes ML, Mercader RC, Taylor MA, Runco J, Desimoni J. Assessment of natural radioactivity levels and their relationship with soil characteristics in undisturbed soils of the northeast of Buenos Aires province, Argentina. *J Environ Radioact* 2012;105:30-9. doi: 10.1016/j.jenvrad.2011.09.014
26. Bolca M, Sac MM, Cokuysal B, Karali T, Ekdal E. Radioactivity in soils and various foodstuffs from the Gediz River Basin of Turkey. *Radiat Meas* 2007;42:263-70.
27. Santawamaitre T, Malain D, Al-Sulaiti HA, Matthews M, Bradley DA, Regan PH. Study of natural radioactivity in riverbank soils along the Chao Phraya river basin in Thailand. *Nucl Instr Meth A* 2011;652:920-24. doi: 10.1016/j.nima.2010.10.057
28. Mohery M, Baz S, Kelany AM, Abdallah AM. Environmental radiation levels in soil and sediment samples collected from floating water from a land runway resulting from heavy rains in the Jeddah region, KSA. *Radiat Phys Chem* 2014;97:16-24. doi: 10.1016/j.radphyschem.2013.10.007
29. El Samad O, Baydoun R, Nsouli B, Darwish T. Determination of natural and artificial radioactivity in soil at North Lebanon province. *J Environ Radioact* 2013;125:36-9. doi: 10.1016/j.jenvrad.2013.02.010

Sažetak

Radioaktivnost u životnoj sredini južne Srbije na lokacijama na kojima je korišten osiromašeni uran

U napadu na Saveznu Republiku Jugoslaviju 1999. godine NATO-ove su snage koristile municiju s osiromašenim uranom. Od 2002. do 2007. Laboratorij za zaštitu od zračenja i zaštitu životne sredine Instituta Vinča proveo je akciju čišćenja osiromašenog urana na lokacijama Pljačkovica, Borovac, Bratoselce i Reljan. U ovom se radu iznose detaljni rezultati praćenja radioaktivnosti u južnoj Srbiji četiri godine nakon spomenute akcije čišćenja (2011), koji uključuje određivanje gama zračenja u uzorcima zemljišta, vode i biljki te određivanje ukupne alfa i beta aktivnosti u uzorcima vode. Rezultati gama spektrometrije pokazali su prisutnost prirodnih radionuklida: ^{226}Ra , ^{232}Th , ^{40}K , ^{235}U , ^{238}U i proizvedenog radionuklida ^{137}Cs . Za procjenu radijacijskog rizika iz zemljišta izračunati su ekvivalent radija, brzina apsorbirane doze gama zračenja, indeks radijacijskog rizika uslijed vanjskog izlaganja i efektivna doza zračenja. Imajući u vidu da na ispitivanim područjima živi znatan broj ljudi, iznimno je važno da se ondje periodično prati sadržaj radionuklida.

KLJUČNE RIJEČI: *uzorci iz životne sredine; gama spektrometrija; ukupna alfa aktivnost; ukupna beta aktivnost*

CORRESPONDING AUTHOR:

Nataša B. Sarap
University of Belgrade, Institute Vinča,
Radiation and Environmental Protection Department,
Mike Petrovića Alasa 12–14, 11001 Belgrade, Serbia
E-mail: natasas@vinca.rs