

## **ASSESSMENT OF DEPLETED URANIUM IN SOIL: A CONTRIBUTION TO METHODOLOGY**

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## ABSTRACT

Absorbed dose rate and natural radionuclide activity concentration measurements were performed to determine the elevation in natural radionuclides, primarily uranium, caused by use of the ammunition with depleted uranium during NATO strikes at targets in the municipality Hadzici in 1995.

Assessment of the DU presence was based on the quantification of uranium/radium disequilibrium and statistical analysis of the obtained results compared with results from previous studies.

Elevated levels of <sup>238</sup>U with high<sup>238</sup>U /<sup>226</sup>Ra disequilibrium which were recorded at several points as well as the irregular distribution of activity concentrations of <sup>238</sup>U and <sup>226</sup>Ra along the two observed soil profiles implied the possible presence of DU.

The obtained results pointed out on the possible use of described method for assessment of depleted uranium presence in soil at points at which the ammunition with DU had been used. The described method could be used together with methods for quantitativequalitative determination of DU.

**Keywords**: depleted uranium, gamma spectrometry, uranium/radium disequilibrium, soil

#### **1. INTRODUCTION**

Depleted uranium (DU) is the accessory product of the enrichment of the natural uranium often used as efficient weapon against armored vehicles and bunkers.

DU penetrators are highly pyrophoric and, after explosion, up to 75% becomes aerosols that can cause contamination of the environment. The affected areadepends on the physical and chemical properties of the aerosols and the prevailing meteorological conditions [15, 16]. Sansone et al. [12] stated that the highest values of DU activity concentrations appear due to DU dust formed when DU penetrators hit hard objects.

Meinrath et al. [9] reported the isotopic composition of DU (0.21 % <sup>235</sup>U, 0.001 % <sup>234</sup>U and approximately 99.8 % <sup>238</sup>U) and activity concentration of <sup>238</sup>U in gram of DU (12410 Bq) which corresponded to activity of 12.41 Bq mg<sup>-1</sup>. Several authors [5,12] stated that soil concentration of <sup>238</sup>U above 3 mg kg<sup>-1</sup> (with <sup>238</sup>U activity concentration above 40 Bq kg<sup>-1</sup>), could be attributed to DU presence in soil. According to mentioned, fast screening of the potentially contaminated soil by DU should primarily be focused on the levels of <sup>238</sup>U above 40 Bq kg<sup>-1</sup>.

Uranium series of radionuclides should be in equilibrium in stabile, undisturbed soil system but many studies suggest the presence of disequilibrium between <sup>238</sup>U and its progeny <sup>226</sup>Ra, with <sup>238</sup>U/<sup>226</sup>Ra activity ratios below 1 [6,11].Furthermore, introducing of sufficient



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amounts of <sup>238</sup>U in soil by depleted uranium, has to lead to disequilibrium between <sup>238</sup>U and its progeny, particularly <sup>226</sup>Ra. Regarding the fact of natural potential for disequilibrium in uranium series of radionuclides, assessment of DU presence by gamma spectrometry method should be based on the background values recorded in affected areas, whenever it would be possible.

According to UNEP report for Bosnia and Herzegovina [16], approximately 3.3 tons of depleted uranium was introduced into the environment of Bosnia and Herzegovina during NATO strikes in 1995. Under the hypothesis that such quantity of radioactive material had to produce the elevated levels of <sup>238</sup>U in soil as well as to uranium/radium disequilibrium at stricken points, the research of the natural radioactivity around the targets stricken by DU penetrators in municipality Hadzici was performed.

For the purpose of the assessment of DU presence in the soil at observed location, it was necessary to have information about measurements performed before NATO strikes in Bosnia and Herzegovina. In doctoral thesis of Lejla Saracevic [13], author reported the levels of natural radionuclides in soil from municipality Hadzici at 0-15 cm depth that were in ranges: 30 - 40 (mean value 31.4) Bq kg<sup>-1</sup> for <sup>238</sup>U; 40 - 50 (mean value 46.7) Bq kg<sup>-1</sup> for<sup>232</sup>Th; 40 - 50 (mean value 47.6) Bq kg<sup>-1</sup> for<sup>226</sup>Ra and 600 - 700 (mean value 662.2) Bq kg<sup>-1</sup> for <sup>40</sup>K.

Quantification of DU presence by gamma spectrometry is mainly associated with uncertainty caused by the presence of natural uranium which makes it hard to reach a precise distinction between these two concentrations. Moreover, a high measuring uncertainty of gamma spectrometry for <sup>238</sup>U contributes to the uncertainty of obtained results. On the other hand, methods like alpha spectrometry imply destroying of the samples which brings a risk of losing targeted material. In the conditions of low concentrations of targeted material (DU, <sup>234</sup>U, <sup>235</sup>U) in the samples, primarily use of quantitative approach could, therefore, lead to uncertainty of the obtained results.

Research was attempted to make a preliminary assessment of DU presence on the base of quantification of uranium/radium disequilibrium, statistical analysis as well as the results from previous studies.

## 2. MATERIALS AND METHODS

## 3. 2.1. Experimental site

Facility for tank reparation in municipality Hadzici was a target at which the DU penetrators were used during NATO strikes in 1995. Locality was mentioned as one of the four localities in Bosnia and Herzegovina at which the presence of DU was recorded in samples of soil, water, air, moss, cabbage and barks [16]. According to the literature data about limited DU contamination [16], the study was performed in surroundings of targetsstricken by DU penetrators during NATO strikes. Inner area of facility for tank reparation was object of the remediation activities performed after UNEP mission in Bosnia and Herzegovina. The sampling of soil was mainly focused on the area around the point at which the buried tank had been stricken by DU penetrator. The point is placed at the hill in the north-east part of the area. The terrain represents the sediments of the Lower Triassic characterized as poorly permeable rocks of the fractured porosity [19]. Dominant soil is Dystric Cambisol [13]. Experimental work was performed during the years 2009 and 2010.

#### 2.2. Measuring of the absorbed dose-rate

The measurements of absorbed dose rate were performed less than 500 meters of air distance from the ffacility for tank reparation as well as at the points presumed to be targets during NATO strikes. Measuring was performed by dosimeter Berthold Model LB 123 at 1 m above the ground with measurement uncertainty of 10 %. Counting range of the used dosimeter was 0.05  $\mu$ Sv h<sup>-1</sup>-10 mSv h<sup>-1</sup>with energy ranges 30 keV -1.2 MeV and resolution of 0.01  $\mu$ Sv h<sup>-1</sup>. Statistical analysis of the results was performed by using of Microsoft Excel 2010.

## 2.3. Sampling and preparing of soil samples

Surface soil samples were taking from 0-15 cm depth at 12 points, positioned in the Facility for tank repair (F), surrounding villages (G, K) and the hill above the facility (H).

For assessment of the uranium distribution along the two soil profiles, samples of soils from the 5 different depths were used (0-5, 5-10, 10-15, 15-30, 30-60 cm).

Soil samples were cleaned of stones, roots and grasses, and dried at 105 <sup>0</sup>C. Dried samples were homogenized by passing through the sieve with 2 mm diameter [7].

Prepared samples were closed into plastic dishes with standard geometry and volume and kept for 120 days till establishing of the secular equilibrium between  $^{238}$ U and  $^{234}$ Th.

#### **2.4.** Gamma spectrometry measurements

The measurements were performed at coaxial HPGe detector ("ORTEC", p-type, Model "GEM 30P4") with relative efficiency 30 % and resolution 1.85 keV at 1.33 MeV. The detector was placed in 10 cm thick lead shield.



Activity concentrations of the observed radionuclides (<sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K) were calculated from their gamma lines as well as from gamma lines of their progeny. Activity concentrations of <sup>238</sup>U were calculated from descendants: <sup>234</sup>Th (energy 63 keV) and <sup>234m</sup>Pa (energy 1001 keV). Levels of <sup>232</sup>Th were calculated from the energies of <sup>228</sup>Ac (911 and 967 keV) and <sup>208</sup>Tl (583 and 2614 keV). Activities of the <sup>226</sup>Ra were obtained from energies of <sup>214</sup>Pb (295 and 352 keV) and <sup>214</sup>Bi (609 and 1764 keV). Activity concentrations of <sup>137</sup>Cs were measured at energy of 662 keV while <sup>40</sup>K activities were calculated at energy of 1461 keV. Measuring time was 80 000 or 50 000 seconds and the results were expressed in Bq kg<sup>-1</sup> of dry weight. For the uncertain results, the measuring was repeated.

#### 2.5. Assessment of depleted uranium

Assessment of DU presence was performed on the base of recorded  $^{238}$ U/ $^{226}$ Ra disequilibrium in soil samples. Bikit at al. [2] established the hypothesis that DU represents excess of uranium concentration above U/Ra equilibrium. Having regard to the fact that DU presence, from viewpoint of gamma spectrometry, should be considered as increased levels of  $^{238}$ U, the mentioned hypothesis could be defined by simple equation:

$$A\left(^{238}U\right) - A\left(^{226}Ra\right) = A(DU) \ (1)$$

Where A represents activity concentrations of naturally occurring <sup>238</sup>U and <sup>226</sup>Ra as well as the artificial DU (Bq kg<sup>-1</sup>).

In order to avoid the overestimating of DU presence in soil caused by the presence of mentioned natural uranium/radium disequilibrium [6,10,11] as well as the presence of measuring uncertainties, it was necessary to establish the limit value of the  $^{238}$ U/ $^{226}$ Ra activity ratio for assessment of DU presence. Despite the fact that average $^{238}$ U/ $^{226}$ Ra activity ratio reported before war was 0.66 [13], the value of 1.0 calculated from results reported in UNSCEAR 2000 [17], was used as background of the mentioned ratio. By adding of the average uncertainty of  $^{238}$ U/ $^{226}$ Ra activity ratio calculation (0.47), the limit value of the  $^{238}$ U/ $^{226}$ Ra activity ratio of approximately 1.5 was obtained. Therefore, all points with recorded  $^{238}$ U/ $^{226}$ Ra activity ratio higher than 1.5 should be considered as points under the suspicion on DU presence. Mentioned approach was used in order to highlight the suspicious points with more significant level of confidence, despite the fact that such approach could have led to underestimating of DU presence rather than its overestimating.

#### **3. RESULTS AND DISCUSION**

#### **3.1 Dose rate measurements**

The average absorbed dose rate in the area of the Facility for tank reparation in the municipality Hadzici as well as in its surroundings was 149.2 nGy  $h^{-1}$ , with values in the range 123–184 nGy  $h^{-1}$ . The highest values of the dose rate were recorded at hill in surroundings of the facility, in the village Kucice (184 nGy  $h^{-1}$ ), whilst the higher dose rates were recorded in directions north-west and south-east from the facility. The spatial distribution of absorbed dose rates at investigated site is shown on the Figure 1.







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Statistical analysis indicated on the relatively inhomogeneous distribution of the dose rate (SD=16.5). Finding of the lower absorbed dose rates in the inner area of the facility were probably caused by remediation activities performed according to recommends of the domestic and international experts. The higher values of absorbed dose rates recorded at nearby hill as well as at points placed on directions north-west and south-east of the facility led to suspicion of the possible DU presence or the presence of higher levels of other radionuclides in soil. According to Bleise et al. [3], who found that DU particles in aerosol could be widespread in downwind directions, obtained increased absorbed dose along the main wind directions could be due to the presence of DU. However, other authors [15,16] were concluded that such contamination should be limited to diameter about 100 meters of the target. The findings were pointed out on the need for quantitative-qualitative analysis of the soil from selected points.

## **3.2** Results of the surface soil samples (0-15 cm depth)

The average activities of the <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K in soil at observed points corresponded to the world average [17], despite the fact that particular results and statistical analysis showed high variability of results, especially for <sup>40</sup>K. Obtained results for soil samples showed very low positive correlation coefficient (R=0.08) between activities of <sup>238</sup>U and <sup>226</sup>Ra. Moreover, results of <sup>238</sup>U/<sup>226</sup>Ra activity ratios as well as recorded <sup>238</sup>U activity concentrations in observed samples were significantly higher then those recorded at the same area before 1995 [13].

According to mentioned hypothesis [2],<sup>238</sup>U excess above <sup>226</sup>Ra level represents the approximative activity concentration of DU under assumption of <sup>238</sup>U/<sup>226</sup>Ra equilibrium. Finding of the low positive correlation between concentrations of <sup>238</sup>U and <sup>226</sup>Ra (R=0.085) as well as the irregular uranium/radium activity ratios pointed on the disequilibrium between these radionuclides probably caused by the presence of DU. Because of the reasons explained above (section 2.5.) limit value of 1.5 was used for <sup>238</sup>U/<sup>226</sup>Ra activity ratio which were indicating the presence of DU.As comparison, values presented in the UNSCEAR report gave the roughly estimated average value of <sup>238</sup>U/<sup>226</sup>Ra activity ratio in soil of 1.03 [17].The points with <sup>238</sup>U/<sup>226</sup>Ra activity ratio above the limit value of 1.5 were: K3, H1, H2, H3, H4, and H6 (Table 1). Mentioned points were not objects of remediation and reparation activities.

# Table 1. Activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K (Bq kg<sup>-1</sup>) in samples of soil (0-15 cm depth) with <sup>238</sup>U/<sup>226</sup>Ra activity ratios at observed points

Point	$^{238}U$	<sup>232</sup> Th	<sup>226</sup> Ra	$^{40}K$	$^{238}U/^{226}Ra$
G	$50.2 \pm 7.5$	$75.2\pm2.0$	$76.2\pm2.0$	$1086.4\pm88.4$	$0.7\pm0.1$
K1	$74.6 \pm 15.1$	$68.8\pm2.4$	$49.9\pm4.1$	$983.7\pm80.3$	$1.5\pm0.4$
K2	$79.5 \pm 16.1$	$65.8\pm2.2$	$64.3\pm4.2$	$979.2\pm79.6$	$1.2\pm0.3$
K3	$75.2 \pm 14.9$	$60.3\pm4.0$	$41.2\pm3.5$	$917.2\pm75.1$	$1.8\pm0.5$
H1	$63.6 \pm 12.7$	$58.7\pm2.2$	$28.8\pm3.7$	$881.5\pm72.5$	$2.2 \pm 0.7$
H2	$46.8\pm9.5$	$57.3\pm4.8$	$27.8\pm4.0$	$777.8 \pm 65.8$	$1.7\pm0.6$
H3	$80.1 \pm 16.0$	$67.1 \pm 4.3$	$32.0\pm3.9$	$964.1\pm78.7$	$2.5\pm0.8$
H4	$60.2 \pm 11.9$	$67.5\pm3.9$	$31.9\pm4.0$	$928.3\pm76.2$	$1.9\pm0.6$
H5	$50.2 \pm 10.3$	$65.9\pm4.3$	$33.5\pm3.3$	$956.3 \pm 79.4$	$1.5 \pm 0.5$
H6	$59.3 \pm 11.5$	$56.6\pm2.1$	$25.9\pm3.5$	$818.4\pm68.1$	$2.3\pm0.8$
F1	$50.4 \pm 4.9$	$58.0\pm2.8$	$48.6\pm2.0$	$842.2\pm70.3$	$1.0 \pm 0.1$
F2	$49.1 \pm 6.8$	$22.7\pm2.5$	$41.0\pm2.1$	$175.3 \pm 18.5$	$0.8\pm0.2$
Mean	61.8	60.3	41.75	859.2	1.6
Min	46.8	22.7	25.85	175.3	0.7
Max	80.1	75.2	76.21	1086.6	2.5
SD	12.8	13.1	15.6	231.0	0.4

Approximate estimation of DU mass concentration could be based on the fact that specific activity of  $^{238}$ U in DU projectile is 12.41 Bq mg  $^{-1}$ [9].Estimating (Equation 1) gave the values of DU contribution to the total  $^{238}$ U concentration activities in range 19 – 48.1 Bq kg  $^{-1}$  at the above mentioned points. According to relation between mass and activity concentration of  $^{238}$ U in DU [15], these values corresponded to mass concentrations of DU in range 1.5–3.9 mg kg  $^{-1}$  of soil.

Estimated mass concentrations of natural <sup>238</sup>U content in soil at mentioned points (K3, H1, H2, H3, H4 and H6) were approximately equal to radium activities and were in range 25.9 - 41.2 Bq kg<sup>-1</sup> which corresponded to mass concentration of natural <sup>238</sup>U between 2.0 - 3.2 mg kg<sup>-1</sup>. This was in accordance with values reported by UNEP mission in Bosnia and Herzegovina [16], which recorded uranium concentrations at uncontaminated points that were in range 1.3-4.8 mg kg<sup>-1</sup> of soil. Total calculated <sup>238</sup>U contents as sum of the natural



 $^{238}$ U and DU at observed points were in range 3.7 – 7.1 mg kg<sup>-1</sup> that still corresponded to natural occurrence (300 µg kg<sup>-1</sup> – 11.7 mg kg<sup>-1</sup> soil) of  $^{238}$ U in soil [18].

Results for the other observed points were not pointed on the presence of DU. Levels of observed radionuclides at some points indicated on the different origin of the soil (F2), as evidence of performed reparation and remediation activities on terrain.

#### **3.3.** Results of the soil profiles (0-60 cm depth)

Investigation of the radionuclide distribution along soil profile (0-5, 5-10, 10-15, 15-30, 30-60 cm depth) was performed at two points selected on the base of distance from the stricken tank.

While there was no information about soil status in terms of its cultivation during the war in Bosnia and Herzegovina (population which lived in the Hadzici during the war, already left their households), vertical distribution of <sup>137</sup>Cs along soil profiles was used for distinction of the cultivated and undisturbed soil. Soil profile 1 showed distribution of <sup>137</sup>Cs typical for cultivated soils while the shape of <sup>137</sup>Cs distribution in the soil profile 2 was more convenient to undisturbed soils [1,4,20]. On the base of those findings, the profiles were classified as cultivated (soil profile 1) and undisturbed (soil profile 2) soils. The comparative distributions of the <sup>238</sup>U, <sup>226</sup>Ra and <sup>137</sup>Cs concentration activities along soil profiles are shown in figure 2 and figure 3.



Fig. 2. Levels of <sup>238</sup>U, <sup>226</sup>Ra and <sup>137</sup>Cs (Bq kg<sup>-1</sup>) in cultivated soil (soil profile 1)



Fig. 3. Levels of <sup>238</sup>U, <sup>226</sup>Ra and <sup>137</sup>Cs (Bq kg<sup>-1</sup>) in undisturbed soil (soil profile 2)

Recorded  $^{238}$ U levels in surface soil layers were decreasing to the depth of 15 cm with slightly increased uranium levelsat depth of 15-30 cm in both profiles. Decreasing trend was further recorded in deeper soil layers (30-60 cm) of the cultivated soil, while the increasing of  $^{238}$ U level was recorded at the same depth of the undisturbed soil.

The most interesting finding was equilibrium between  $^{238}$ U and  $^{226}$ Ra activities at 30 - 60 cm depth in cultivated soil. That finding pointed out on the depth of 30-60 cm in cultivated soil which DU reached to. It was supported by the findings of the high  $^{238}$ U/ $^{226}$ Ra activity ratio in the first observed soil layer of cultivated soil (1.77), compared with the second layer (1.56) and the third one (1.38). Mentioned ratio in layer 30-60 cm depth was 1.02 (Fig. 4.).





## (P1-cultivated soil, P2-undisturbed soil and results from 1990)

The levels of  $^{238}$ U and  $^{226}$ Ra did not reach the equilibrium at the same depth of undisturbed soil (soil profile 2). Increased activities of both radionuclides followed by the lower  $^{238}$ U/ $^{226}$ Ra disequilibrium were recorded deeper layers of the undisturbed soil. The distribution of  $^{238}$ U was relative homogenous along the observed 60 cm depth of undisturbed soil, with generally higher and more

The distribution of  $^{238}$ U was relative homogenous along the observed 60 cm depth of undisturbed soil, with generally higher and more uniform  $^{238}$ U/ $^{226}$ Ra activity ratios (1.34-1.64; figure 5.), which had still existed in the deeper layers. The finding led to conclusion that soluble fraction of DU was probably migrated to the depths below 60 cm. As comparison, UNEP mission in Bosnia and Herzegovina reported maximal migration of DU to the depth of 40 cm [16].

Activity concentrations of  $^{238}$ U and  $^{226}$ Ra showed positive correlation in both profiles with different correlation coefficients between the profiles (R<sub>1</sub>=0.73, R<sub>2</sub>=0.25), with values lower than usual [18]. Recorded disequilibrium between  $^{238}$ U and  $^{226}$ Ra activities, as well as lower correlation coefficients between the levels of these two radionuclides in undisturbed soil, indicated on the possible presence of DU in undisturbed soil as well as in the upper soil layers of the cultivated soil.

Regarding to the fact that analysis of the physical and chemical properties of soil profile samples were not performed, differences in DU migration among soil profiles could not be clearly explained. Differences in leaching rate were recorded by Schimmack et al. [14] after 3 years of studying of DU corrosion and leaching in two soil type columns. They reported the large temporal and inter-column variability of the <sup>238</sup>U leaching and concluded that leaching rates obtained after 3 years should not be used to estimate those in the future.

Observing the radio ecological situation in Kosovo, Sansone et al. [12] recorded the leaching of uranium isotopes down to the depth of approximately 18 cm, eighteen months after NATO strikes of Kosovo. They also highlighted the possible risk of groundwater contamination in the future. Study of the uranium isotopes in water springs in the area of town Hadzici, performed by Vidic et al. [19] was not reported the significant presence of DU. Having regard to the fact that there was no evidence of significant contamination of the drinking water in municipality Hadzici [19], it could be assumed that leachable fraction of deposited DU probably did not reach to the depth of underground water reservoirs. It seemed that the total leachable fraction of deposited DU had been used up on uniform distribution along soil profiles in undisturbed soil, before it reached to the depths of underground reservoirs. Additionally, results obtained for cultivated soil showed significant fixation of the majority of deposited DU in upper soil layers. This hypothesis led to assumption that there was no more DU which could endanger the underground water resources, taking into consideration the time expired from deposition. The assumption would be valuable only in conditions without the changing of the physical and chemical properties of soil that would have had an impact on the solubility and fixation of the deposited DU.

Taking into consideration the fact that study represented the investigation without using the more sophisticated and precise techniques like ICP MS, the obtained results were analysed on the base of approximate approach, with use of current knowledge and available techniques. Despite the fact of the rough values obtained by assessment, these preliminary results represent valuable information for the future investigations. Methods and techniques described in the study could be used for investigations of DU presence at other localities, together with quantitative-qualitative methods for determination of DU.

## 4. CONCLUSION

Obtained results confirm the theses that introduction of the particular substance into environment has to lead to its presence in some parts of the ecological chain. In the case of introducing of DU into environment, appearance of  $^{238}U/^{226}Ra$  disequilibrium in the surroundings of DU ammunition targets could be used for DU assessment.



Despite the fact that recorded levels of  $^{238}$ U did not show significantly increased values,  $^{238}$ U/ $^{226}$ Ra activity ratios, statistical parameters as well as the results from previous studies, indicated the presence of DU at some of observed points. Assessment of the total uranium concentrations with included DU concentrations, is slightly higher than world average value but still under the ange of the natural occurring of  $^{238}$ U.

Soil profile analysis pointed out at the possible DU presence under the first 30 cm depth of cultivated soil, while its presence in undisturbed soil appeared to be deeper than 60 cm depth. Obtained results indicate the need for the further investigations at wider area as well as deeper soil layers with use of the more sophisticated techniques.

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