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## DETERMINATION OF DEPLETED URANIUM IN ENVIRONMENTAL SAMPLES BY GAMMA-SPECTROSCOPIC TECHNIQUES

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### ABSTRACT

The use of depleted uranium (DU) in military operations causes the contamination of the target area due to DU dispersion in the environment. Since the isotopic abundance of uranium in DU is different than that in nature, the only evidence of DU contamination is the disruption of the natural isotopic abundance of  $^{238}\text{U}$  and  $^{235}\text{U}$ . For the investigation of the existence of such a disruption, the accurate determination of  $^{235}\text{U}$  and  $^{238}\text{U}$  is necessary. The Nuclear Engineering Section of the National Technical University of Athens (NES-NTUA) uses high-resolution LEGe detectors and especially developed gamma spectroscopic analysis techniques for the determination of  $^{238}\text{U}$ , and  $^{235}\text{U}$  and subsequently for the isotopic abundance of the uranium isotopes in the sample.  $^{235}\text{U}$  is determined from the analysis of the multiplet photopeak at 186 keV. DU may be detected with this technique provided that the  $^{238}\text{U}$  activity of the DU is higher than ~20% of that of  $^{238}\text{U}$  in the natural uranium of the sample. The analyses of surface soil samples collected well before the dispersion of depleted uranium in Kosovo by NES-NTUA, made evident that Kosovo is an area of high natural background, with a pronounced surface soil variation in natural uranium content, and  $^{238}\text{U}$  activity reaching values as high as  $330 \text{ Bqkg}^{-1}$ . It was also observed that disturbance of radioactive equilibrium among the nuclides of the uranium series is quite often observed due to leaching and weathering, with the ratio of the activities of  $^{238}\text{U}$  and  $^{226}\text{Ra}$  ranging from 0.17 to 5.5. After the military operations in Kosovo, surface soil and vegetation samples as well as DU penetrators were analysed in NES-NTUA. Among the soil samples analysed we found that some of those collected around a DU penetrator crater were contaminated by DU, with  $^{238}\text{U}$  activity exceeding  $2 \text{ kBqkg}^{-1}$ , and ratio of  $^{238}\text{U}/^{226}\text{Ra}$  exceeding the value of 10. In these soil samples, the natural isotopic abundance of the

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uranium isotopes was significantly disrupted. It allowed for an estimation of the isotopic abundance of  $^{235}\text{U}$  in the dispersed DU of about 0.2%, which agrees very well with relevant values found in the literature for DU penetrators. DU in the vegetation samples analysed was below detection limit. The analysis of DU penetrators led to the detection  $^{238}\text{U}$ ,  $^{235}\text{U}$  and their daughters, and to an estimation of the dose-rate due to the gamma-rays emitted from the penetrator, which reached  $6.5 \mu\text{Sv h}^{-1}$  on the surface, sloping to background at the distance of about 40 cm.

Key words: environment, depleted uranium, DU ammunition, DU penetrator, measurements, gamma spectrometry

## 1. Introduction

The use of depleted uranium (DU) penetrators in military operations causes the contamination of the target area, because of the DU aerosol formation during impact of the penetrator on a hard target. DU aerosol is then dispersed into the environment at a distance that strongly depends on various parameters such as environmental conditions, aerosol size distribution, temperature etc. Since both the uranium isotopes  $^{238}\text{U}$  and  $^{235}\text{U}$  found in DU penetrators, are naturally occurring radionuclides, DU is characterized as Technologically Enhanced Naturally Occurring Radioactive Material (TENORM). The determination of DU contamination in environmental samples, such as soil, water and vegetation is difficult, because the DU  $^{238}\text{U}$  and  $^{235}\text{U}$  are also detected in almost all environmental samples, where natural uranium exists as trace element. The isotopic abundance of natural uranium is 99.28%  $^{238}\text{U}$ , 0.72%  $^{235}\text{U}$  and  $^{234}\text{U}$  0.006%. The latter is almost always ignored mainly due to its very low concentration. This abundance is not expected to be disturbed in the soil, unless there is some kind of contamination with depleted or enriched uranium. The only exception has been reported to be the Oclo phenomenon [IAEA, 1975]. The only difference between natural and depleted uranium is in the isotopic abundance, which in case of the DU used in penetrators is 0.2% for  $^{235}\text{U}$ , 0.001% for  $^{234}\text{U}$  and the remaining (approx 99.8%)  $^{238}\text{U}$  [McClain, 2001]. Thus, the only indication of DU contamination in a sample is the statistically justified disruption of natural isotopic abundance of the uranium detected in it. One of the analytical techniques used for the qualitative and quantitative determination of uranium is gamma spectroscopy. It is a non-destructive analytical technique for the determination of gamma emitting radionuclides, which in general does not require any sample treatment prior to the analysis. Both  $^{238}\text{U}$  and  $^{235}\text{U}$  may be determined by gamma spectroscopic techniques. Their analysis is difficult, because of their extremely long half-lives which result to low photon emission rates, and the fact that it is difficult to detect and identify the photons they emit. Therefore special detectors and techniques should be used. Moreover, the content of the natural uranium isotopes and their decay products in soil prior to contamination provide very useful information for the determination of DU contamination in the soil. In this paper we presented:

1. Data for the natural radioactivity of the surface soil of Kosovo obtained prior to the military operations in 1999, and compared with similar data from Greek surface soils also collected prior to the operations.

2. A technique developed in NES-NTUA for the gamma spectroscopic determination of DU contamination in environmental samples.
3. Results of the analyses of surface soil and vegetation collected in Yugoslavia after the military operations in 1999, from DU targeted areas, for the determination of DU contamination.
4. Measurements of the gamma ray dose rate close to a DU penetrator.

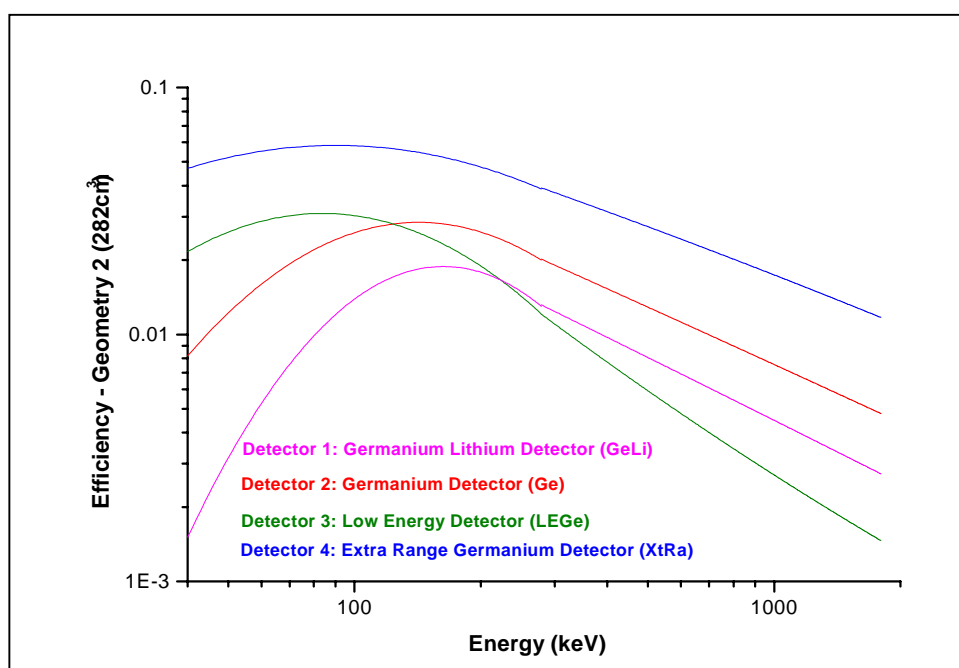
## 2. Gamma spectroscopic determination of $^{238}\text{U}$ and $^{235}\text{U}$ in soil samples

Determination of uranium in soil is in most cases synonymous to  $^{238}\text{U}$  determination, since the assumption of natural isotopic abundance of the uranium isotopes in the soil is always made, unless there is suspicion for enriched or depleted uranium contamination. Moreover, since  $^{238}\text{U}$  does not emit significant photons that can be used for its gamma spectroscopic determination, it is usually determined through its daughter products in equilibrium. The daughter products that are most often used are  $^{234}\text{Th}$  and  $^{234\text{m}}\text{Pa}$ . The latter emits 1001.03 keV photons with very low yield (0.59%), and therefore it cannot be used for the accurate determination of  $^{238}\text{U}$  in low activity samples. On the other hand,  $^{234}\text{Th}$ , due to its short half-life (24 days), is almost always in equilibrium with  $^{238}\text{U}$  in soil samples, and it emits low energy photons with low yield at the energies: 63.29 keV (3.8%), 92.35 keV (2.72%) and 92.78 keV (2.69%). Moreover, the photons at 92.35 keV and 92.78 keV are very close, resulting to a multiplet photopeak, which is very difficult to be analysed. For this reason these two peaks are not often used, though there are cases where an approximation is made and the multiplet photopeak is treated as a single one with yield 5.41% [Roessler, 1979]. The analysis of the low energy photons at 63.29 keV requires the use of special detectors, with high efficiency in the low energy region below 200 keV, such as Low Energy Germanium (LEGe) detectors, or Extended Range Germanium (XtRa) detectors. For such low energy photons, the count-rate during the  $\gamma$ -spectroscopic analysis is highly affected by the intense self-absorption of the photons inside the sample, especially in large volume samples, depending on the sample geometry, the type of material, the material packing density and the photon energy. Note that the self-absorption of photons in the material is generally different than that of the efficiency calibration standard. A method has been recently developed in NES-NTUA [Anagnostakis, 1995a], for the quantitative determination of radionuclides emitting low-energy photons. This method takes into account the difference in the self-absorbing properties between the calibration source material and the analysed material, introducing an efficiency correction factor. Correlations valid for photon energies between 46-186 keV and soil densities  $\sim 0.5\text{-}2\text{ grcm}^{-3}$ , developed and reported [Anagnostakis, 1998; Petropoulos, 2001], have been applied for the determination of the linear attenuation coefficient  $\mu(\rho, E)$  that is needed for the efficiency correction factor calculation. The  $^{235}\text{U}$  activity in soil samples is not very often reported, mainly due to the difficulties in its determination.  $^{235}\text{U}$  emits photons with energies: 143.76 keV (10.96%), 163.33 keV (5.08%), 185.72 keV (57.2%) and 205.31 keV (5.01%). Due to its long half-life ( $7.04 \cdot 10^8$  y) and the very low photon emission rate of all these photons in environmental samples, the only photons that could be used for the determination of  $^{235}\text{U}$  are those at the energy of 185.72 keV. The problem with the analysis of these photons is due to the fact that  $^{226}\text{Ra}$ , a daughter of  $^{238}\text{U}$  that emits 186.25 keV photons, co-exists with  $^{235}\text{U}$  in environmental samples, and the photons which these two nuclides emit, result to a multiplet photopeak at  $\sim 186$  keV, difficult to be analysed into its two components. If radioactive

equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$ , and natural isotopic abundance for uranium are assumed, the contributions of the photons emitted from  $^{235}\text{U}$  and  $^{226}\text{Ra}$  recorded to this multiplet photopeak may be calculated, thus allowing for an estimation of the activities of the two isotopes [Khien, 1981]. In the case that radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  cannot be assumed,  $^{235}\text{U}$  may be determined from the 185.72keV photons, provided that the  $^{226}\text{Ra}$  activity has been previously determined from the  $^{226}\text{Ra}$  daughters in equilibrium. However, in this case the inaccuracy in the determination of  $^{235}\text{U}$  depends on the inaccuracy in the determination of  $^{226}\text{Ra}$ .

For the analysis of  $^{238}\text{U}$  and  $^{235}\text{U}$  in environmental samples, such as soil, two detectors are used in NES-NTUA:

1. A Low Energy Germanium (LEGe) detector, with 341 eV fwhm at 5.9 keV, and 530 eV at 122 keV, calibrated in the energy region 0-200 keV.
2. An Extended Range Germanium (XtRa) detector, with 1003 eV fwhm at 122 keV and 107% relative efficiency, calibrated in the energy region 0-2000MeV.



**Figure 1. Full energy peak efficiency curves for bulk geometry (282cm<sup>3</sup>) of NES-NTUA detectors.**

The efficiency calibration curves of the Ge detectors of NES-NTUA for the sample geometry used are presented in Figure 1. The advantage of LEGe and XtRa detectors for this kind of analysis is prominent. During the gamma spectroscopic analysis other important nuclides of natural radioactivity, such as  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  and  $^{40}\text{K}$  as well as any artificial gamma-emitting radionuclides in the soil are also determined. Plastic, cylindrical boxes of 282 ml are filled up with the examined samples. The boxes are hermetically sealed and covered with a film of epoxy resin, to limit, as far as possible, the escape of radon from the boxes which ensures secular equilibrium of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  with their decay products,

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[Simopoulos, 1987]. All specimens are analysed at least three weeks after the boxes have been sealed. The  $^{226}\text{Ra}$  radioactivity is derived from the weighted mean of the activities of two photopeaks of  $^{214}\text{Pb}$  (295.2, 352.0 keV) and of three photopeaks of  $^{214}\text{Bi}$  (609.3, 1120.3, 1764.5 keV). In the case of  $^{232}\text{Th}$  two photopeaks of  $^{228}\text{Ac}$  (338.4, 911.1 keV) and the photopeaks of  $^{212}\text{Pb}$  (238.6 keV) and  $^{208}\text{Tl}$  (583.1 keV) are used in the same way. The radioactivity of  $^{40}\text{K}$  is obtained from the single photopeak of this isotope at 1460.75 keV. Further details on the calibrations performed, the  $\gamma$ -spectroscopic analysis and the quality assurance procedures followed, may be found elsewhere [Simopoulos, 1987].

The technique used in this work for the determination of  $^{235}\text{U}$  is based on the analysis of the multiplet photopeak at  $\sim 186$  keV using the LEGe detector calibrated in the low energy region 0-200keV, which allows for sufficient number of channels per keV (e.g.  $\sim 20$  channels / keV). The spectrum analysis software used was SPUNAL (SPectrum UNix AnaLysis), an in-house developed computer code capable of analysing complex gamma spectra with up to 10 component multiplets, using a modified Marqadt algorithm [Petropoulos, 2001]. SPUNAL code is suitable for the detection and the analysis of close-lying and/or overlapping peaks, which may constitute a multiplet, such as the one at  $\sim 186$  keV and the one at  $\sim 92.5$ keV from the adjacent 92.38 keV and 92.80 keV photons of  $^{234}\text{Th}$ . Furthermore, SPUNAL performs self-absorption corrections in the low energy region.

From the results obtained so far, it was found that the multiplet photpeak at  $\sim 186$  keV may be analysed into its two components, at 185.75 and 186.25keV, leading to the direct determination of  $^{235}\text{U}$  and  $^{226}\text{Ra}$  respectively, in the cases where the statistics of the multiplet photopeak at  $\sim 186$  keV was satisfactory. It should be noted that sufficient statistics was obtained with analysis times of about  $5 \cdot 10^5$ s.

In conclusion, for every sample analysed:

- The  $^{238}\text{U}$  activity was determined indirectly from the 63.29 keV photons emitted by its decay product  $^{234}\text{Th}$ .
- The  $^{226}\text{Ra}$  activity was determined both indirectly from its decay products in equilibrium and directly from the 186.25 keV photons in the case where the multiplet photopeak at  $\sim 186$  keV was analysed into its two components. It is already reported [Petropoulos, 2001] that both methods agree within the total errors of the measurements.
- The  $^{210}\text{Pb}$  activity was determined using its 46.54 keV photons.
- The spectrum of a soil sample in the energy region 0-200keV is presented in Figure 2.

### 3. Determination of DU contamination in soil samples

Since the only evidence of DU contamination in soil samples is the disruption of the natural isotopic abundance of  $^{238}\text{U}$  and  $^{235}\text{U}$ , an accurate determination of these nuclides should be performed. Taking into consideration the natural isotopic abundance of the uranium isotopes (99.2% for  $^{238}\text{U}$ , and 0.72% for  $^{235}\text{U}$ ), and their half-lives, the activity ratio of natural uranium is calculated to  $^{238}\text{U}/^{235}\text{U} = 21.7$ . In the cases where this ratio is statistically significantly higher than 21.7, there is evidence that the analysed sample is contaminated with DU; in these cases the isotopic abundance of the uranium in the contaminated soil may be determined. The analysis of the multiplet photopeak at  $\sim 186$  keV for a soil sample contaminated with DU, and a natural soil sample with radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$ , are presented in Figure 3.

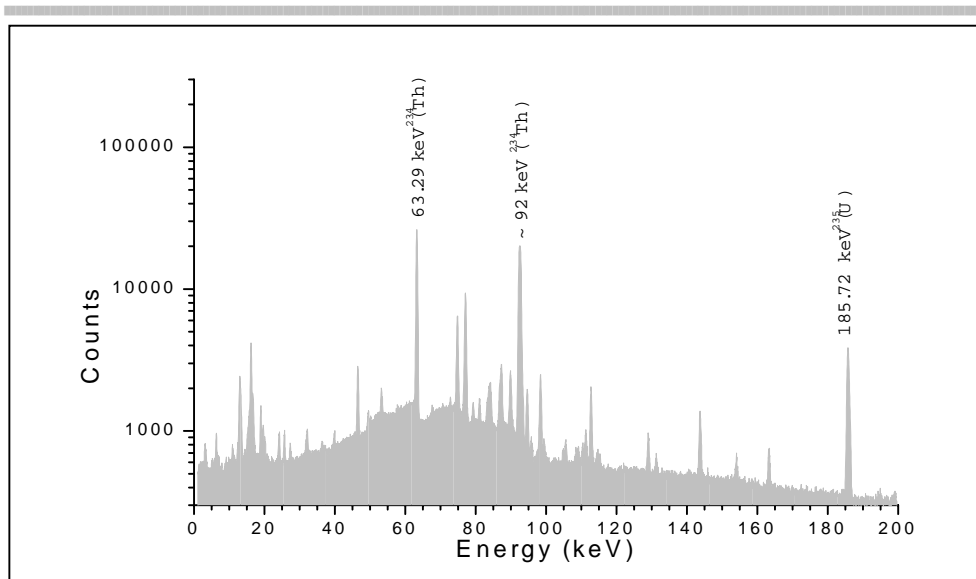


Figure 2.  $\gamma$ -spectrum of DU contaminated soil sample in the energy region 0-200keV.

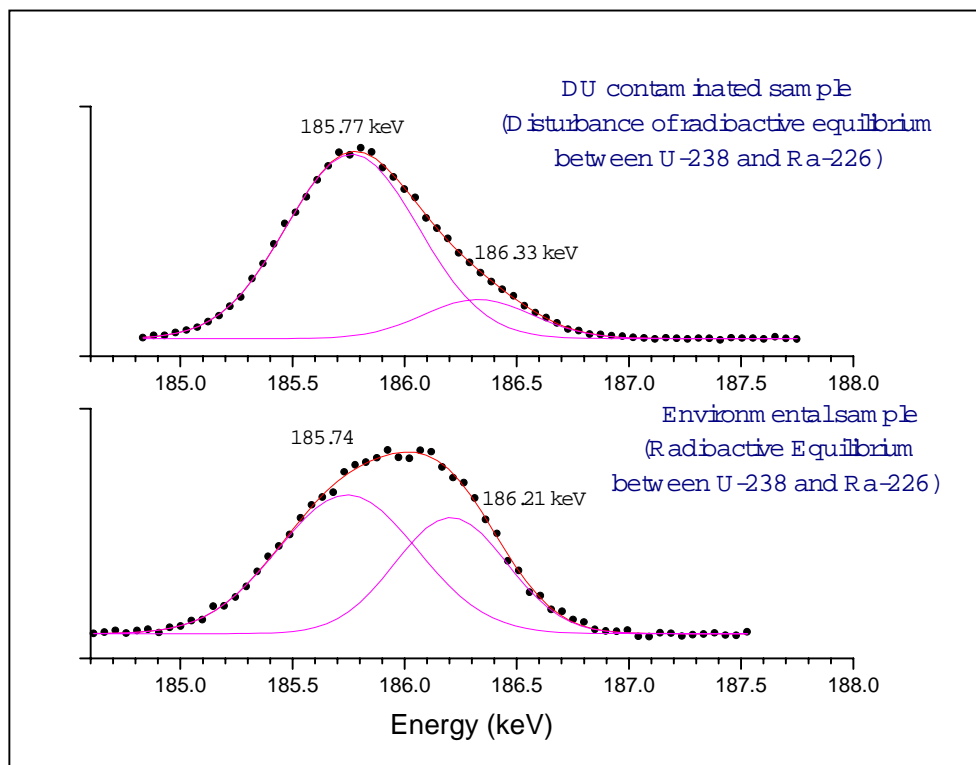


Figure 3. Analysis of the multiplet photopeak at ~186keV: (a) Soil contaminated with DU, (b) Soil not contaminated, with radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$ .

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For the determination of the DU content of the soil, the following assumptions have to be made: (a) natural isotopic abundance of uranium existed in the soil, prior to contamination, and (b) the DU penetrators are the only source of DU contamination of the soil. Furthermore, one of the following two assumptions has also to be made:

1. The isotopic abundance of the DU contaminant is known. A typical value for DU used in kinetic penetrators is 0.2% for  $^{235}\text{U}$  and  $\sim 99.8$   $^{238}\text{U}$  [McClain, 2001]. In this case the fractions of  $^{238}\text{U}$  and  $^{235}\text{U}$  activities that should be attributed to the natural uranium of the soil and the DU contaminant, may be calculated.
2. Radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  existed in the soil prior to DU contamination. In this case, the  $^{238}\text{U}$  and  $^{235}\text{U}$  activities in the soil prior to DU contamination are determined from the  $^{226}\text{Ra}$  activity. Since both,  $^{238}\text{U}$  and  $^{235}\text{U}$  activities in the soil prior and after contamination are known, the activities of  $^{238}\text{U}$  and  $^{235}\text{U}$  in  $\text{Bqkg}^{-1}$ , as well as their isotopic abundance in the DU contaminant are determined. One should be very careful when performing these calculations, since radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  in the soil is quite often disturbed.

In the present research the second assumption was adopted; furthermore the isotopic abundance of  $^{235}\text{U}$  in the DU was calculated and it did not show a statistically significant difference from the nominal value of 0.2%.

One important issue when performing this kind of analyses is the limit of detection of DU in the analysed samples. For this determination, the relatively low emission rates of the 63.29 keV ( $^{238}\text{U}$ ) and 185.72 keV ( $^{235}\text{U}$ ), which result to significant uncertainties in their determination, should be taken into consideration. In the case of the LEGe detector of NES-NTUA, the standard errors in the determination of  $^{235}\text{U}$  (185.72 keV) and  $^{238}\text{U}$  (63.29 keV) are of the order of 2% and 4% respectively, resulting to a standard error of the order of 5% for the determination of the ratio  $^{238}\text{U}/^{235}\text{U}$ . Taking this into consideration, DU contamination in the soil may be detected, provided that the  $^{238}\text{U}$  activity of the DU is higher than  $\sim 20\%$  of that of  $^{238}\text{U}$  in the natural uranium of the sample.

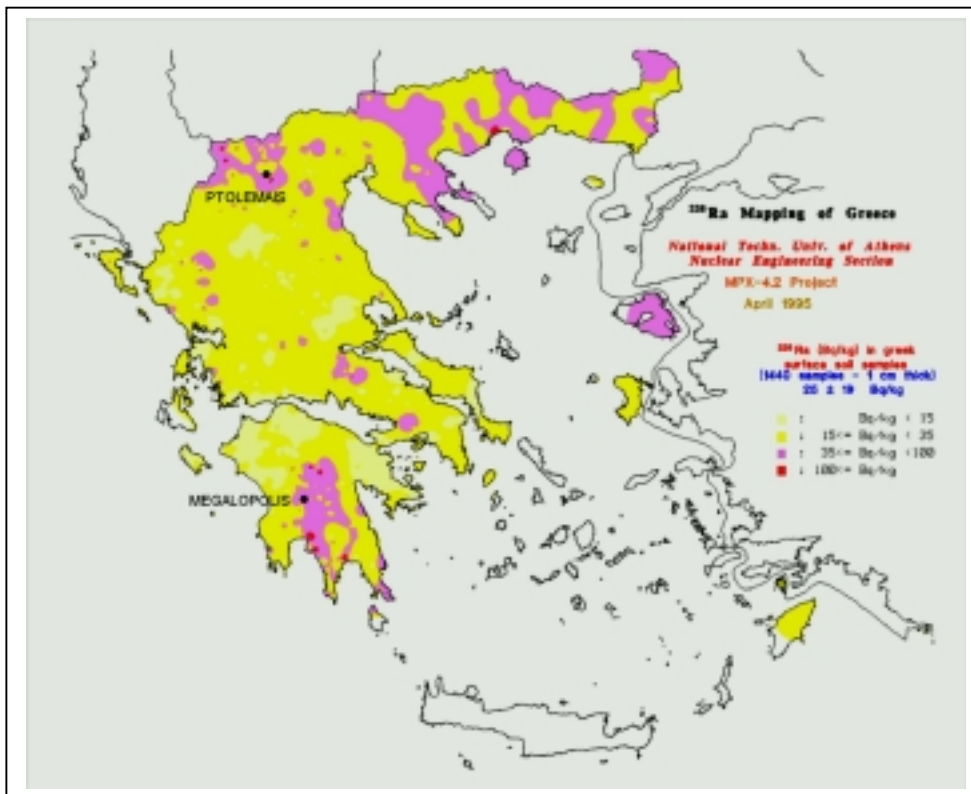
#### **4. Natural radioactivity of pre-war surface soil samples from the southern of Yugoslavia**

NES-NTUA has established a scientific co-operation with the Institute of Nuclear Sciences Vinča of Yugoslavia since 1996, with a mutual aim to study the natural radioactivity in Yugoslavia. In the framework of this project the study of the natural radioactive background of Kosovo and the southern part of Serbia started beginning 1998, about one year before the military operations in the area. Surface soil and rock samples collected from the area, by the scientists of the Vinča Institute, were analysed at NES-NTUA using the above gamma spectroscopic techniques. The results obtained from these analyses are presented in Table 1, from which the following results are drawn:

Natural radioactivity in the investigated locations is in some cases elevated, with  $^{238}\text{U}$  and  $^{226}\text{Ra}$  activities exceeding the value of  $300 \text{ Bqkg}^{-1}$  and  $^{232}\text{Th}$  exceeding  $200 \text{ Bqkg}^{-1}$ . These values are high, compared to those of Greek surface soils, as it is deduced from Figure 4, where the mapping of  $^{226}\text{Ra}$  radioactivity of Greek surface soils is presented [Anagnostakis, 1995b].

**Table 1. Pre-war soil samples analysis results.**

Sample Code	Location	Activity in Bqkg <sup>-1</sup> ± error 90%					
		<sup>238</sup> U	<sup>226</sup> Ra	<sup>210</sup> Pb	<sup>232</sup> Th	<sup>40</sup> K	<sup>238</sup> U/ <sup>226</sup> Ra
MS088	Servia	326± 9%	205± 1%	199± 10%	210± 1%	1694± 1%	1.59
MS089	Gornja Stubla	164± 7%	190± 0.3%	132± 4%	243± 0.5%	1546± 0.4%	0.86
MS090	Kosovo	28± 8%	32± 1.3%	50± 6%	32± 1.3%	354± 1%	0.88
MS091	Kosovo	219± 5%	162± 0.3%	147± 4.4%	171± 0.5%	1333± 0.4%	1.35
MS092	Kosovo	325± 3%	336± 4%	362± 3%	231± 0.6%	1554± 0.6%	0.97
MS094	Kosovo	37± 6%	29± 0.6%	33± 9%	31± 0.9%	357± 0.6%	1.28
MS095	Kosovo	300± 5%	249± 0.4%	303± 4%	87± 0.8%	1531± 0.6%	1.20
MS096	Kosovo	57± 7%	55± 0.7%	51± 10%	87± 0.8%	65± 3.4%	1.04
MS097	Kosovo	20± 5%	28± 0.9%	42± 8%	28± 1%	244± 1.0%	0.71
MS098	Kosovo	32± 4%	26± 0.8%	50± 4%	31± 1.0%	228± 0.8%	1.23
MS099	Gornja Stubla	326± 5%	234± 0.3%	265± 3%	233± 0.6%	1618± 0.5%	1.39
MS100	West Serbia	32± 4%	-	35± 7%	-	386± 0.6%	-
MS101	West Serbia	30± 6%	-	48± 6%	-	310± 1.4%	-
MS102	Gornja Stubla	325± 2%	266± 4%	323± 2%	160± 0.6%	1668± 0.4%	1.22
MS103	Gornja Stubla	320± 3%	208± 0.4%	237± 4%	164± 0.6%	1585± 0.4%	1.54
MS104	Gornja Stubla	6.5± 4%	-	6± 9%	-	3± 3.4%	-
						Mean value :	1.2
						Standard Deviation :	0.27



**Figure 4. Mapping of <sup>226</sup>Ra in greek surface soils.**



**Table 2. Post-war samples analysis results.**

Sample Code	Sample type	Location	Activity in Bqkg <sup>-1</sup> ± error 90%					
			<sup>238</sup> U	<sup>235</sup> U	DU ( <sup>238</sup> U)	<sup>226</sup> Ra	<sup>210</sup> Pb	<sup>40</sup> K
MS115	vegetation	Plackovica-Vranje	---	---	---	---	34 ± 9%	551 ± 3%
MS116	soil	Plackovica-Vranje	23 ± 11%	---	---	---	34 ± 9%	276 ± 4%
MS117	vegetation	Plackovica-Vranje	9 ± 6%	---	---	---	51 ± 16%	482 ± 3%
MS118	vegetation	Plackovica-Vranje	---	---	---	---	76 ± 11%	203 ± 3%
MS119	vegetation	Plackovica-Vranje	---	---	---	---	23 ± 21%	592 ± 3%
MS120	vegetation	Plackovica-Vranje	---	---	---	---	58 ± 5%	250 ± 4%
MS121	soil	Plackovica-Vranje	53 ± 6%	---	---	---	67 ± 5%	735 ± 1%
MS122	soil	Bratoselec	1000 ± 2%	19 ± 7%	828 ± 7%	172 ± 11%	197 ± 10%	-
MS123	soil	Bratoselec	737 ± 3%	15 ± 9%	544 ± 9%	193 ± 9%	194 ± 9%	1132 ± 2%
MS124	vegetation	Bratoselec	---	---	---	---	26 ± 10%	323 ± 3%
MS125	vegetation	Bratoselec	---	---	---	---	42 ± 10%	305 ± 3%
MS126	soil	---	---	---	---	---	39 ± 12%	357 ± 4%
MS127	vegetation	Bratoselec	---	---	---	---	2 ± 25%	308 ± 3%
MS128	vegetation	Bratoselec	49 ± 7%	---	---	---	49 ± 11%	1030 ± 1%
MS129	soil	Bratoselec	2417 ± 1%	37 ± 6%	2248 ± 6%	169 ± 15%	201 ± 12%	-
MS130	soil	Plackovica	66 ± 6%	3 ± 10%	---	54 ± 13%	87 ± 8%	-
MS131	soil	---	1854 ± 1%	29 ± 3%	1714 ± 3%	140 ± 7%	168 ± 7%	1168 ± 1%
MS143	soil	Plackovica	50 ± 8%	---	---	41 ± 1%	46 ± 8%	657 ± 3%
MS144	soil	Vrbovac	47 ± 7%	---	---	---	63 ± 8%	644 ± 3%
MS145	soil	Mesto B	40 ± 7%	---	---	40 ± 1%	49 ± 6%	658 ± 3%

**Table 3. <sup>238</sup>U/<sup>226</sup>Ra in environmental samples.**

Origin of soil samples	Sample Size	<sup>238</sup> U/ <sup>226</sup> Ra activity
Greek surface soils	70	0.17 – 5.5
Soils from Yugoslavia (not contaminated with DU)	16	0.71 – 1.59
Soils from Yugoslavia (contaminated with DU)	4	3.8 – 14.3

**Table 4. <sup>235</sup>U abundance in DU contaminated soils and DU penetrators**

Sample	<sup>238</sup> U in Bqkg <sup>-1</sup>	<sup>235</sup> U in Bqkg <sup>-1</sup>	<sup>235</sup> U abundance in DU contaminated soil (%)	<sup>235</sup> U abundance in DU penetrator (%)
MS122	1000	19	0.304	0.217
MS123	734	15	0.320	0.178
MS129	2417	37	0.242	0.206
MS131	1854	29	0.247	0.210
			Mean	0.203

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Radioactive equilibrium among the nuclides of  $^{238}\text{U}$  series is, in some cases, significantly disturbed, with the  $^{238}\text{U}/^{226}\text{Ra}$  ratio in the range 0.71–1.6 and the  $^{210}\text{Pb}/^{226}\text{Ra}$  ratio in the range 0.74–2.0. These results are consistent with similar results from Greek surface soils, where the ratio  $^{238}\text{U}/^{226}\text{Ra}$  ranges from 0.17 to 5.5 [Anagnostakis, 1998]. This disturbance may be attributed to the weathering of rocks and the activity of water on the soil.

## 5. Analysis of post-war samples from DU target areas

After the military operations in Kosovo, the co-operation with Vinča Institute continued in the determination of DU contamination in environmental samples, and especially in soil and vegetation from DU target areas in the southern Serbia. A total of 20 samples - 11 soil and 9 vegetation samples - were collected by scientists from Vinča Institute, and analysed in NES-NTUA using the LEGe and XiRa detectors and the gamma spectroscopic techniques developed for the determination of DU in environmental samples. From the results that are presented in Table 2 it is concluded that a total of four soil samples were contaminated with DU, with the DU content exceeding in one case  $2200\text{ Bqkg}^{-1}$ , in terms of  $^{238}\text{U}$ . The activity ratios  $^{238}\text{U}/^{226}\text{Ra}$ , for the four contaminated samples, are presented in Table 3, together with the same ratios for not contaminated surface soils from Greece and from Yugoslavia, for comparison purposes. From Table 3 the following conclusions may be drawn:

- DU contaminated samples are generally highly enriched in  $^{238}\text{U}$ , compared to  $^{226}\text{Ra}$ . The  $^{238}\text{U}/^{226}\text{Ra}$  ratio in the four DU contaminated soil samples ranges from 3.8-14.3, well above unity.
- $^{238}\text{U}/^{226}\text{Ra}$  for non-contaminated soil samples may be as high as 5.5, because of radioactive equilibrium disruption. For this reason, high values of the ratio  $^{238}\text{U}/^{226}\text{Ra}$  should not be considered as an evidence of DU contamination.
- Values of  $^{238}\text{U}/^{226}\text{Ra}$  higher than about 5 should be normally considered as an indication of DU contamination.

Further to the confirmation of DU contamination in four soil samples, the isotopic abundance of uranium in the soil and in the DU fallout was determined, under the assumption of radioactive equilibrium between  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in the samples, prior to contamination. From Table 4, where the isotopic abundance for the four samples are presented, it may be concluded that the  $^{235}\text{U}$  abundance in the DU penetrators has a mean value of about 0.2%, which agrees with similar values previously reported [McClain, 2001].

DU penetrators collected from target areas were also analysed by  $\gamma$ -spectroscopy. The only isotopes detected were  $^{235}\text{U}$ ,  $^{238}\text{U}$  and their daughters. Furthermore, the dose-rate due to the gamma rays emitted from the penetrator, was  $6.5\ \mu\text{Svh}^{-1}$  on the surface, sloping to background at the distance of about 40cm (Figure 5).

## 6. Conclusions

From the methods and the results presented, the following conclusions may be drawn:

- The pre-war natural radioactive background in some parts of the southern Yugoslavia is elevated, with the  $^{238}\text{U}$  activity in surface soil exceeding in some cases  $300\text{Bqkg}^{-1}$ . Thus, high  $^{238}\text{U}$  activity in surface soils from these areas does not necessarily mean DU contamination.

2. Gamma spectroscopy may be used for the determination of DU contaminated environmental samples, provided special detectors and techniques are used. One such technique, developed in NES-NTUA for this determination, is presented.
3. The lower limit of detection for DU contamination in environmental samples strongly depends on the natural uranium content of the analysed samples. With this technique DU may be detected only if  $^{238}\text{U}$  in the DU contaminant is higher than about 20% of the naturally occurring  $^{238}\text{U}$  in the sample.
4. High  $^{238}\text{U}/^{226}\text{Ra}$  ratio in environmental samples does not necessarily imply DU contamination, and it could be due to the disruption of radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$ . However, according to the results obtained so-far, significant disruption of radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  in an analysed sample, with  $^{238}\text{U}/^{226}\text{Ra} > \sim 5$ , may be an indication of DU contamination.
5. In a few of the soil samples collected at DU target areas in Yugoslavia after the military operations in 1999, that were analysed in NES-NTUA, high levels of DU contamination was detected.

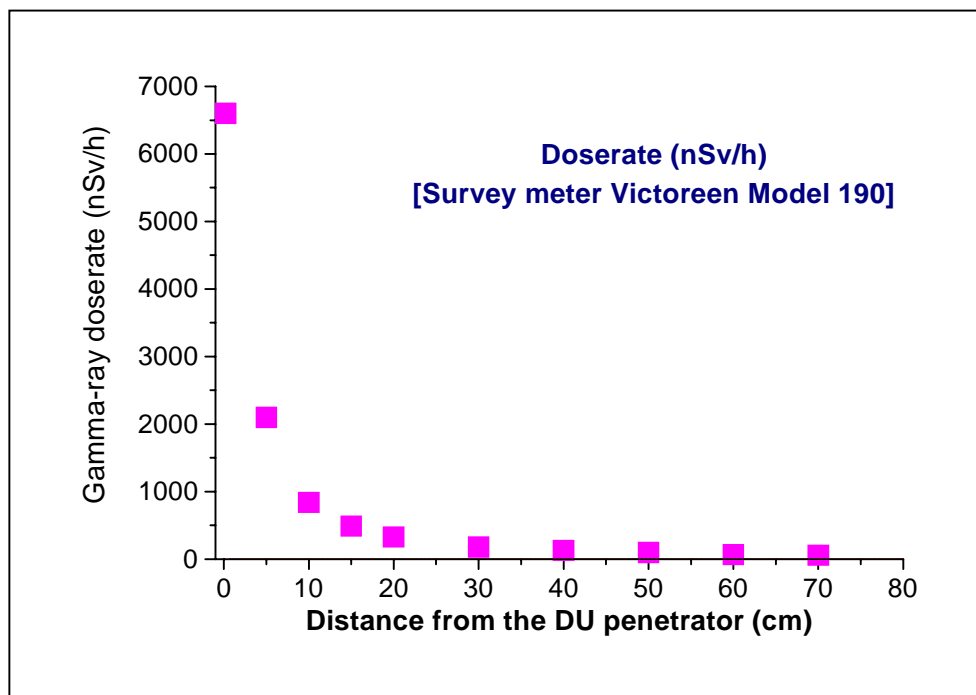


Figure 5.  $\gamma$ -ray dose rate due to DU kinetic penetrator.

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