Depleted Uranium in Kosovo
Results of a survey by gamma spectrometry on soil samples

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ABSTRACT

The possible presence of depleted uranium (DU) in the soil of former Yugoslavia (FRY) after the 1999 conflict raised great public opinion concerns all over the world. The so-called Balkan-syndrome is often linked with DU-contamination. An excellent compilation of data about DU and its possible impact on health and environment can be found in the UNEP report[1] and publications from the Swedish SSI[2]. Unfortunately, very few systematic and reliable data on the possible DU concentrations are until now available. Some of these rare data, mainly available on the web, are obtained with inadequate measurement techniques and their "interpretation" is often biased by prejudices of some (pressure)groups[3]. To clarify the situation, a systematic survey was started in the summer of 2000 as a collaborative effort between Ghent University (Physics Laboratory) and the Belgian Ministry of Defence (Medical Service). From 50 sites selected all over Kosovo, 150 soil samples were measured in the laboratory with a high resolution gamma-spectrometer. Some sites (14) were explicitly selected based on military information on the use of DU munitions there. After careful analysis we can conclude that there is no indication of any DU contamination on these 50 sites with a 15 Bq minimal detectable activity; this corresponds approximately to 1mg DU in a typical sample (100 - 150 gram). Based on our data, it is very unlikely that military personnel in Kosovo is facing health hazards from radiation due to DU exposure.

INTRODUCTION: Depleted Uranium

DU is a by-product of the nuclear fuel production; when the fissile ^{235}U is separated from the natural uranium during the enrichment process, a resulting waste product is DU, containing app. 99.74% ^{238}U. Chemically, it behaves like natural U, but its activity is less (only 60 to 75% of the natural U, depending on the enrichment process). It is also possible that DU is originating from the reprocessing of burned nuclear fuel elements; in that case some traces (0.01% – 0.1%) of ^{236}U can be expected[4]. Speculations on the presence of relevant amounts of Pu in that type of DU are until now never been confirmed.
Uranium (and DU) is a heavy metal with chemical toxicity, like many other heavy metals (Pb, Hg etc.). Typical activity for natural uranium (without daughter products) is 26 Bq/mg; DU (of type 1) has 19.4 Bq/mg (following the data in the CEA-report[4])
Due to its very high density, DU is used as counterweight in ships, planes (747) and rockets.

From the military point of view DU-penetrators in bullets give very high impact forces for piercing armour plates. DU powder is also pyrophoric; it can ignite spontaneously at temperatures above 600°. The combination of those two properties makes DU-ammunitions very lethal for tanks and soldiers inside.

Fig. 1 PGU-14 bullet

Tonnes of DU were used during the military operations in Kosovo, mainly under the form of API (Armour Piercing Incendiary) bullets (fig. 1 is type PGU-14, 30mmx173mm, containing a 292 gram DU penetrator). During a typical attack 3 US A-10 Warthog airplanes use 9 to 18 kg DU in 150-300 rounds of DU ammunition[1]. When a such a bullet hits a target, it is crushed into small particles an dust; most of the dust is deposited within 100 m from the impact.

ORGANISATION OF THE SURVEY

To obtain reliable data on the possible DU soil contamination in Kosovo, a systematic survey was organized as a collaborative effort between Ghent University (Physics Laboratory) and the Belgian Ministry of Defence (Medical Service).
Kosovo is of special interest for us because Belgian soldiers are still there in the frame of NATO-force. In the press and public opinion much concern raised about the presumed role of uranium in different kinds of illness of some soldiers after their duty in Kosovo: the so-called Balkan syndrome.

During the sample collection campaign in the summer of 2000, 50 places all-over Kosovo were visited by a military team under supervision of one of the authors (ML).
Sites were randomly chosen but they included military positions, open fields, road sides, and locations **near** places targeted with classical or DU-containing ordnances.
The term “near” means that the samples are taken outside the impact area, but within a 5 to 15 meter radius around the impact point (if known exactly).
The correct position of the DU sites was based on KFOR information providing us with the exact locations and the amounts fired.
We have reliable information that on 14 sites, indicated on the map with a blue point, DU ammunitions have been used. The collection campaign was performed independently from NATO authorities; photographic evidences from the collection sites were taken.
In figure 2 a map with the collecting points is presented.

On each site 3 samples were collected at depths between 0-10 cm, 20-30 cm and 40-50 cm and stored in sealed 100 ml PE bottles. This way, we have an extra quality control on the sampling procedure, by following the depth profile for different isotopes (see next paragraph).

Fig. 2: map of the collecting sites in Kosovo: regular sampling sites (red), DU sites (blue)

EXPERIMENTAL METHODS

To trace the presence of DU in the soil samples, we have chosen the gamma-spectroscopic measuring method to discriminate between the natural uranium (and the other natural isotopes in the soil) and DU. If we consider the decay scheme of uranium[5] (fig. 3) we can see that high resolution gamma spectrometry is an relatively easy and suitable tool for this purpose.
In the decay of natural uranium (99.27% ^{238}U, 0.718% ^{235}U) we have an equilibrium between the U-mother (half-life 4.47 x 10^9 year) and the Pb and Bi daughter isotopes far in the decay process; the typical gamma radiation of ^{214}Pb (242, 295 and 352 keV) and ^{214}Bi (609 and 1120 keV) have good and well-known intensities and can easily be measured with a germanium detector.
In the case of DU however, the decay process starts with pure ^{238}U, without any daughters isotopes (removed in the enrichment process). The long half-life of ^{234}U (246 000 y) (see the DU-limit indication on fig. 3) is a real barrier for the formation of ^{214}Pb and ^{214}Bi because any DU production started only after 1940! Only the ^{234}Pa and ^{234}Th isotopes are in equilibrium with ^{238}U after a few months. Fortunately, we have a typical but weak 1001.03 keV line in the decay of ^{234m}Pa that can be used for the selective determination of DU. We use a sensitive low background set-up with a heavily shielded P-type Ge-detector (34% efficiency, 1.75 keV resolution fwhm) (fig. 4) coupled to an InSpector2000 (Canberra Ind.) with DSP and trapezoidal pulse-shaping (12µs rise time, 0.8µs flat top). The detector is calibrated with NIST (formerly National Bureau of Standards) sources SRM 4276 (mixed-radionuclide Eu-Sb) and SRM 4353 (Rocky Flats soil).

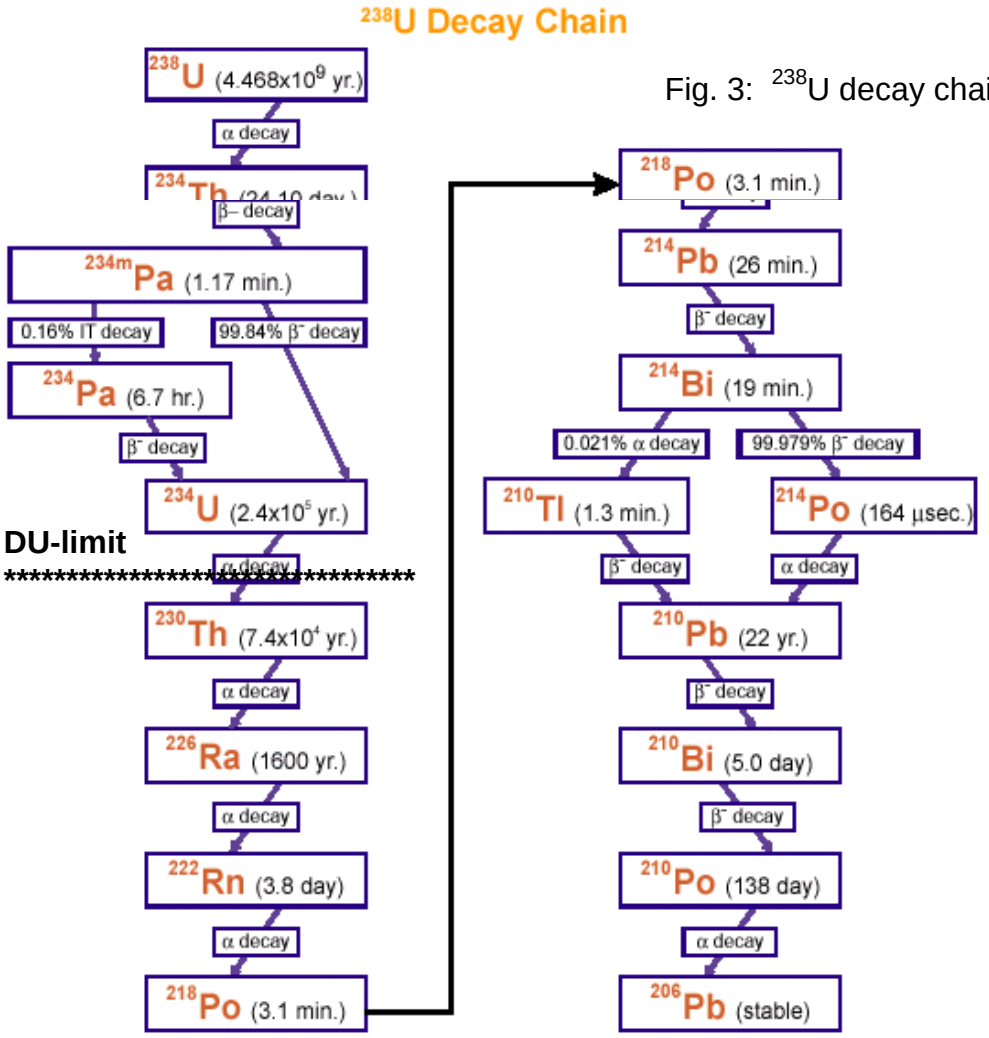

Fig. 3: ^{238}U decay chain[5]

In a 4096 channel spectrum, covering the energy range from 50 to 1500 keV, we can measure in each sample all the gamma-emitting isotopes from the natural uranium and thorium series, ^{40}K, DU, and the ^{137}Cs (from the Chernobyl fall-out).

All those isotopes can be measured via relatively high energy gamma lines (238 to 1461 keV), without problems with self-absorption; no special source preparation is required.

Fig. 4: a sample in front of the detector inside the shielding

Absorption and detector efficiency problems are the reason why the low energy lines (98 keV from the ^{234m}Pa decay and 62 and 92 keV lines from the ^{234}Th decay) are not really useful for the determination of DU.
As one can see in the spectrum in fig 5, lower trace, the mean background of our set-up in the 1000 keV region is very low (2.2 counts/channel in 10 000s). The only drawback is the small emission probability of the 1001 keV-line; we use the value 0.835(2) %, based on the recent review article by Nzuruba[6]. One should be very careful about the ^{234m}Pa intensities used in different catalogues and software packets; old values of 0.59 for this 1001 keV line can be found! Also in the case of the second, very weak 766.36 keV line from the ^{234m}Pa decay the situation is not clear: one can find values between 0.294% (in the R.L. Heath catalogue[5] and in the Canberra NuChart-based software) and 0.315(3)% in Nzuruba[6]. The background in the 766 keV region is also much higher than in the 1001 keV region; consequently, we use the 766 keV line only for control and confirmation purposes and all our calculations are based on the 1001.03 keV line.

One should always remember that also the natural uranium in the soil gives a very small 1001 keV line. Strictly spoken, our DU measurements are based on the excess ^{234m}Pa activity in the 1001 keV line. However, with the normal uranium concentrations in the 25 Bq/kg region, this “natural” contribution falls into the background and no 1001 keV line is detected in absence of DU (as one can see in the lower trace of fig. 5)

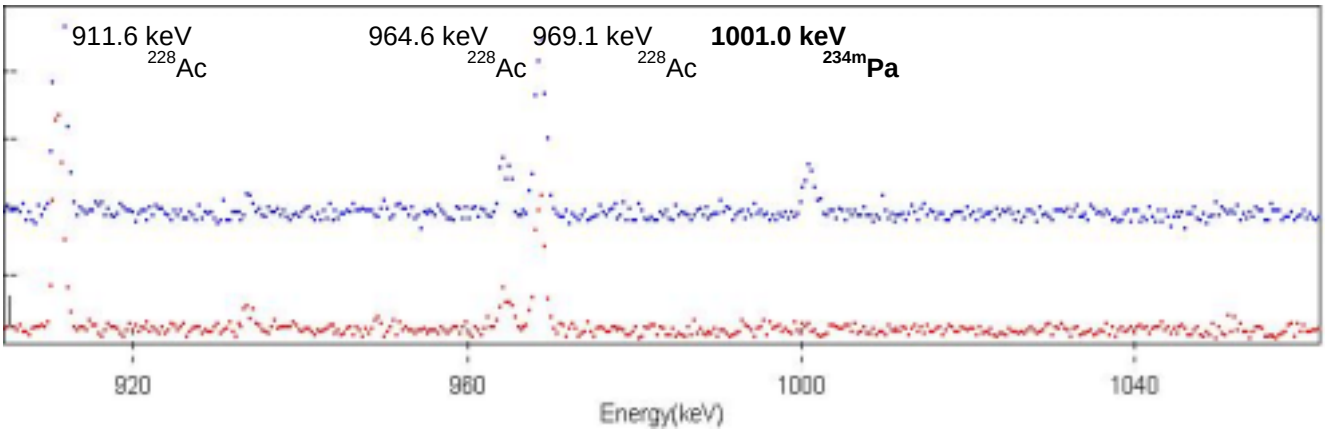

Fig. 5: High energy region of 2 spectra (measuring time 40 000s)
lower trace: a typical spectrum of a Kosovo soil sample (52A, 133 g); no DU detected
upper trace: a calibration spectrum with 2.4 milligram DU added to a similar soil sample

The 3 samples collected at each location must give coherent data for the uranium, the thorium series and the ^{40}K (homogeneous distribution in the soil); the 661.6 keV line from ^{137}Cs is mainly expected in the top layer sample. Because of this, our spectroscopic method has internal controls on the quality of the sample taking procedure.

MDA

An important figure of merit is the sensitivity of our set-up or what is the minimal detectable activity (MDA). A usable approach[7,8] defines MDA as the maximum amount of activity for a particular nuclide that could be present and remain undetected in a real spectrum. The MDA expression takes into account detector parameters (resolution, photo peak efficiency and peak-to-compton ratio), crystal shielding (background), isotope properties and measuring time in the following combination:

$$MDA(E_1) = \frac{A_m}{e_f(E_1).f.t_m}\left(\sqrt{2bR(E_1) < B(E_1) > + A_m^2/4} + A_m/2\right)$$ with following notations:

A_m : is the reciprocal of the fractional error (e.g. 20% means A_m=5)
$e_f(E_1)$: is the absolute detector efficiency at E_1 in the given source geometry
f : is the branching ratio or relative intensity
t_m : measuring time
b : shape factor; approx. 2 for symmetrical gaussian peaks
$R(E_1)$: is the energy resolution (fwhm) at E_1, expressed in channels
$<B(E_1)>$: is the mean background in a channel in the E_1 peak region; it is composed of the natural background contribution from the shielding and from the Compton contributions from higher energy lines

In the case of the 1001 keV line from ^{234m}Pa the MDA is affected negatively by the very small intensity (0.00835); on the other side the low background of our set-up in the 1000 keV region is favourable.
The so calculated 20% MDA value is 20 Bq for an 11 h. measuring time and 15 Bq for the longer 18 h measuring time.

To confirm our calculated MDA-values, calibration samples containing a well-defined amount of DU as a hot particle in the centre of the bottle have been prepared.
The upper trace in fig. 5 is part of the spectrum from a control soil sample doped with 2.4 ($\pm$0.2) milligram DU. Three ^{228}Ac lines (from the natural ^{232}Th-series) are very resembling, but the 1001 keV line from ^{234m}Pa is exclusively and unambiguously present in the calibration spectrum. Using the uranium (and ^{234}Pa) conversion factor[1] of 12.27 Bq/mg, a value of 30 ($\pm$3) Bq is expected. The measured activity value after analysis with the Genie 2000 software[9] is 33 ($\pm$7) Bq, in excellent agreement with the expected value.
A second calibration source containing only 0.9 ($\pm$0.2) milligram DU was used in a very long 100 000s test measurement with satisfying results.

Summarizing, we have proven by MDA-calculations and experimentally, that we are able to detect DU-particles in the milligram region in an untreated soil sample by gamma spectrometry in a reasonable measuring time.

RESULTS AND CONCLUSIONS

In ***none*** of the 150 measured samples a significant trace of the 1001 keV line was found. Based on our MDA-considerations (and the experimental confirmation with calibration sources), we can state with good confidence that there is no DU present at our 50 sampling points in Kosovo, with MDA values as low as 15 Bq (corresponding to milligram DU amounts). Some samples, taken near places where DU-ammunitions were used, have been re-examined very carefully with extra long measuring times (27.8 h), always with negative results.
Based on these data, it is very unlikely that military personnel currently deployed in Kosovo are facing health hazards from DU exposure. Our research does not address the situation in other regions of the FRY and our conclusions are based on 150 samples; it is possible that on other places traces of DU can be found. ***However, it is very unlikely that, if DU-particles should be widely spread over the region, not one single particle in the milligram range is present in our samples.***
Our conclusions are in agreement with the (preliminary) findings of the Portuguese research group [12].

As supplementary information, we have calculated the following results for some other isotopes:

Uranium series: mean value 25 Bq/kg (based on ^{214}Pb and ^{214}Bi (5 lines))
Thorium series: mean value 37 Bq/kg (based ^{228}Ac, ^{212}Pb and ^{208}Tl (5 lines))
^{40}K: mean value 370 Bq/kg (based on the 1460.8 keV line)
The mean values for the natural isotopes in Kosovo are quite similar with those found in the south of Belgium[10].

The artificial ^{137}Cs, resulting from atmospheric fall-out (nuclear bomb testing, Chernobyl accident) is easily measured via the intense 661.6 keV line; the mean value for the top layer is 53 Bq/kg. This figure is difficult to compare with the values in Belgium[11], obtained by in-situ spectrometry and expressed in Bq/m^2, but on some places the concentrations in Kosovo are considerably higher than in Belgium.

It is worth while to note that we have found values for ^{137}Cs between practically zero and heights between 200 and 600 Bq/kg in some top layers samples. People using portable contamination monitors (measuring gross β and γ- radiation without sufficient energy resolution) should be very cautious not to attribute increased count rates on some places to DU; a locally higher ^{137}Cs concentration is often more plausible.

References

1. UNEP/UNCHS Balkans Task Force (BTF), *The potential effects on human health and environment arising from possible use of depleted uranium during the 1999 Kosovo conflict* (October 1999)
2. Snihs, J.A. & Akerblom, G., *Use of depleted uranium in military conflicts and possible impact on health and environment*. *IRPA-10 proceedings (CD-ROM)*, P-1b-S2, Hiroshima, Japan (2000).
3. Compare for example http://www.gulflink.osd.mil/du_index.htm_ with http://www.llrc.org/depleted_uranium.htm
4. Uranium, uranium appauvri, effets biologiques (CEA report in french) http://www.cea.fr/actualite/archives.asp?id=77
5. Heath, R.L. *Gamma ray spectrum catalogue (4th edition)* 832-835 (1998). http://id.inel.gov/gamma/data1.html or new data (2000) on 238U: http://id.inel.gov/cgibin/byteserver.pl/gamma/pdf/u238.pdf
6. Nzuruba, A.C., *Evaluation of absolute gamma ray emission probabilities in the decay of ^{234m}Pa*. Nucl. Instr. Meth. A424, 425-443 (1999).
7. Cooper, J.A., *Factors determining the ultimate detection sensitivity of Ge(Li) gamma-ray spectrometers*. Nucl. Instr. Meth. 82, 273-277 (1970)
8. Uyttenhove, J.A. and Poffijn, A., *Considerations on the use of high resolution gamma spectrometry in environmental measurements*. Rad. Prot. Dos. 34, 207-209 (1990).
9. Canberra Industries, Genie-2000 Spectroscopy software, V1.4 (1999).
10. Uyttenhove, J.A., Pommé, S. & Van Waeyenberge, B., *Measurement of natural gamma radiation in Belgium by means of high resolution in-situ spectrometry*. IRPA-10 proceedings (CD-ROM), P-1a-14, Hiroshima, Japan (2000)
11. Uyttenhove, J.A. et al., *Survey of the ^{137}Cs contamination in Belgium by in-situ gamma spectrometry, a decade after the Chernobyl accident*. Health Phys. 73-4, 644-646 (1997).
12. Preliminary Report (in Portuguese) http://www.mct.pt/relatoriopreliminar Final version, also in English http://www.itn.pt/English/en_index.html