DECONTAMINATION OF CAPE ARZA (MONTENEGRO) FROM DEPLETED URANIUM

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1. INTRODUCTION

On May 30, 1999, NATO A-10 aircrafts attacked Cape Arza, a very attractive touring area on peninsula Lustica, at the entrance of Boka Kotorska Bay, in Montenegro. They fired anti-armour rounds with penetrators made of depleted uranium.

Such an armour-penetrating round has a length of 173 mm and a diameter of 30 mm. The bullet has an aluminium case (jacket) and inside it a conical DU penetrator. The length of the penetrator itself is 95 mm, and the diameter of its base is 16 mm. The penetrator weight is 292 g.

According to the data reported by NATO (NATO, 2001), the total number of rounds fired against Cape Arza was 480. As to the data on combat mix of the A-10 aircraft gun, 300 (UNEP, 2001) or 400 (UNEP, 2001; FAS) of these rounds where with DU penetrators, and the rest with a classical charge. This means that Cape Arza was contaminated with 90 or 120 kg of DU, or with a radioactivity of (3.5 - 4.7)×10⁹ Bq.

Depleted uranium is a waste product of the process of uranium enrichment in ²³⁵U isotope, for use in nuclear reactors or in nuclear weapons. The isotopic composition of depleted uranium is (Harley et al., 1999): (99.7 – 99.8) % of ²³⁸U, (0.2 – 0.3) % of ²³⁵U, 0.001 % of ²³⁴U, and only traces of ²³⁴Th, ²³⁴Pa and ²³⁴Th. If traces of the isotopes ²³⁶U, ²³⁹Pu and ²⁴⁰Pu are also present, as it is the case with DU from Cape Arza (UNEP, 2002), the depleted uranium is obtained by reprocessing of spent nuclear reactor fuel. The activity concentration of depleted uranium is 39.42·10⁶ Bq/kg. Most of it comes from ²³⁸U and its decay products ²³⁴Th and ²³⁴Pa which are in radioactive equilibrium (12.27·10⁶ Bq/kg per each of them), and the less part from ²³⁵U and ²³¹Th (0.16·10⁶ Bq/kg per each) (UNEP, 1999), while the activity concentration of ²³⁶U, ²³⁹Pu and ²⁴⁰Pu is below 100 Bq/kg (UNEP, 2001).

Therefore, the total activity of one whole DU penetrator is 11.51·10⁶ Bq. The penetrator emits α, β and γ-radiation. The total energy of α-radiation emitted from depleted uranium in unit time amounts to 11% of that in the ²³⁸U-series of decays in radioactive equilibrium. For β-radiation this ratio is 42 %, and for γ-radiation 1.4 %.

Although depleted uranium is considered a low-radioactive waste, its specific activity of 39.42·10⁶ Bq/kg is 2.4·10⁵ times higher than maximum natural specific activity of ²³⁸U in the ground of Montenegro, which amounts to 165 Bq/kg (Vukotich et al., 1997).

In the year 1991, Montenegro declared itself as an ecological state. Therefore, the Government of Montenegro has decided to start financing decontamination of Cape Arza.
from depleted uranium ammunition, the first-ever undertaking of this kind in the world as far as we know. After some perspective investigation at the site, which has shown a radiation level up to 50 times higher than the local natural background, the project of decontamination was finished in December 2000, and preparatory works carried out during January 2001.

The systematic survey and decontamination of Cape Arza started in February 1, 2001, before NATO made public their data about use of DU ordnance in Yugoslavia (NATO, 2001), and before the report of UNEP Depleted Uranium Assessment Team (UNEP, 2001) was published. The campaign lasted to June 15 of the same year, and continued during April and May 2002, with engagement of the team of 12 domestic professionals (7 civilians and 5 army officers) and about 10 people in logistics. The area of about 37,000 m² has been surveyed, and DU contamination found on 16,000 m² and cleaned up. The work will be continued after summer season, and hopefully finished before the end of this year. An area of about 20,000 m² in the hinterland of Cape Arza, where our coarse preliminary survey has shown presence of depleted uranium ammunition, will be systematically surveyed and decontaminated.

2. INSTRUMENTS

For measurements in the field we had at disposal one BERTHOLD's portable radiation measuring system TOL/F and several domestic contamination monitors: KOMO-TL (or RMK-10) and KOMO-TM.

1. The TOL/F portable system consists of a basic unit LB 1320 and a set of various hand probes. Each of the system combinations can work as a dosimeter or as a counter, and has audible sound-signals accompanying radiation detection.

   a.) If the ionization chamber probe LB 1321 is connected to the basic unit, the TOL/F becomes the radiation dosimeter. The chamber has a cylindrical shape, with an active zone 10 cm in length and 2.6 cm in diameter, which is filled with an air-equivalent gas mixture. The energy range is 10 keV to 7 MeV. In the low dose rate range $0.1 \mu Sv/h – 10 mSv/h$ the probe operates in the proportional range, and in the high dose measuring range $10 mSv/h – 100 Sv/h$ in the ionization chamber range.

   b.) With the hand probe LB 1231 connected to the basic unit, the TOL/F becomes $\beta$ and $\gamma$-contamination monitor. This probe has the $\beta\gamma$-detector LB 6357, which is a large-area proportional counter tube filled with xenon gas. Neither refilling nor flushing is required. The counter tube window is made of a titanium foil of 5 mg/cm² density. The effective area of the tube is 160 cm². Because the $\beta\gamma$-detector cannot distinguish between these types of radiation, the displayed count rate is the sum of $\beta$ and $\gamma$-radiation induced pulses per unit of time.

   c.) If the hand probe LB 1232 is connected to the basic unit, the TOL/F operates as $\alpha$ and $\beta$-contamination monitor. The probe has a large-area butane-filled proportional counter tube. An extremely thin foil (0.3 mg/cm²) seals the counting chamber. Because this window foil may easily get damaged even during the tube refilling, and especially by the sharp objects in the field (the detector must be held very close to the ground due to a very short range of $\alpha$-radiation), we decided not to use this detector for searching for DU contamination at Cape Arza.

2. The KOMO-TL is a contamination monitor manufactured by the Institute of Nuclear Sciences "Vinca". The detector is a Geiger-Müller tube of the pancake type S8 (Russian made), designed to register contamination from $\beta$ and $\gamma$-ray emitting radionuclides.
The detector window is made of a thin mica foil (14 – 17 µm), and protected against damage by a metal net. The effective window area is 27 cm². The instrument can give a sound alarm each time a radiation event is detected.

3. The KOMO-TM is also a contamination monitor manufactured by the Institute "Vinca". Its detector is a halogene Geiger-Müller tube, with a mica window of 6 cm² effective area. The instrument is able to give audible sound-signals.

Before starting the field measurements we performed some laboratory experiments to test and compare capacities of these instruments for detecting penetrators and DU contamination.

I.) First we checked these measuring devices by a calibration β-radiation source ⁹⁰Sr+⁹⁰Y (plane source with surface of 160 cm²). This source was chosen because the radioisotope ⁹⁰Y emits the β-radiation with maximum energy $E_{β}^{max} = 2280.1$ keV, which is almost the same as the maximum energy of the most intensive β-radiation from depleted uranium ($E_{β}^{max} = 2268.9$ keV of the ²³⁴mPa isotope) (Chu et al., 1999). Each of the detectors was positioned 1 cm above the center of the source.

The experiment has shown that the KOMO-TL counter and the TOL/F system with βγ-probe have approximately the same efficiency if both the detectors are exposed only to the part of source surface equal to the sensitive area of the KOMO-TL detector. The KOMO-TM counter has in this experimental arrangement 2.5 to 3 times lower efficiency than these instruments, but one should have in mind that its sensitive window area is about 4 times smaller than the corresponding one of the KOMO-TL detector. In the case when the whole surface of the source is unshielded, efficiency of the TOL/F βγ-monitor is 5 times higher than that one of the KOMO-TL.

II.) Using the quasipoint radiation sources ¹³⁷Cs and ²²⁶Ra, shielded with 4 mm of aluminum, we compared detection efficiencies of these instruments for γ-radiation. Source-detector distance was 1 cm. Both KOMO instruments have shown similar efficiency, while TOL/F with βγ-probe had 2.5 to 3 times higher efficiency compared to them.

III.) In order to get an idea how deep in the ground of Cape Arza we are able to detect the presence of DU penetrators with these instruments, we performed the following experiments.

Red soil (terra rossa), with a density of 1.24 g/cm³, is typical for Cape Arza. At the bottom of a large metallic vessel we positioned one whole penetrator horizontally. Then, over the penetrator we added red soil layer by layer, keeping the soil density at the constant value of 1.24 g/cm³, and measuring radiation with each one of the instruments, positioned above the center of the penetrator, until reaching the background value of readout. Afterwards, we did the same experiment but with the penetrator in a vertical position, with its sharp top downward. During these measurements, the windows of all instruments were protected by strong plastic foil, just as they were later used in the field at Cape Arza. This foil decreases detection efficiency of β-radiation from ⁹⁰Sr + ⁹⁰Y source by 20 %, but it is an indispensable protection of the contamination and damage of the detector window by the sharp rocks and remains of cut vegetation. The foil practically does not decrease detection efficiency of γ-radiation.
The TOL/F $\gamma$-probe has shown the lowest sensitivity to depleted uranium. It detects DU penetrator up to a depth (8 - 16) cm in the ground, depending on its position. This is the reason why we decided not to use this probe for the dosimetric survey of Cape Arza.

The instruments KOMO-TL and KOMO-TM have almost equal sensitivities to depleted uranium, and they enable locating the penetrator up to a depth of (14 – 18) cm, depending on its position inside the ground. However, the KOMO-TL monitor has advantage for terrain survey because of the larger sensitive area of its detector. On the other hand, the KOMO-TM monitor, due to the small sensitive surface of its detector, appears as the instrument of choice when, inside the given impact place, the penetrator buried in the ground should be located following a narrow radioactive trace that remains behind it.

In the above-described experiments the TOL/F $\beta\gamma$-monitor proved itself as the instrument with the best capacities for identifying DU contamination and penetrators in the field, because it enables location of penetrators up to a depth of (20 – 25) cm in soil, and because its detector has the largest sensitive area. But this monitor is the heaviest and largest of all, and therefore inappropriate for carrying and sweeping it close to broken ground. This is why we decided to use it only for checking presence of radioactive contamination at a given place, and for verifying the successfulness of the decontamination performed.

IV.) We have also made an experiment to determine the range of $\beta$-radiation from $^{90}$Sr + $^{90}$Y in the red soil from Arza. We found that already the 1 cm thick layer of soil above this plane source makes the measuring instruments give count rates that are on the background level for this type of soil.

Accordingly, detection of $\beta$-radiation during a field survey enables discovering points with ground surface contamination by depleted uranium dust, while detection of the $\gamma$-radiation enables locating the uranium penetrators and their fragments buried in the ground.

3. EXPERIENCE FROM CAPE ARZA

Cape Arza is a rocky terrain with scarce thin layers of soil, which is mostly of terra rossa type, rarely brown or humus soil. The cape is covered with shrubs and bushes, and trees are only few. Terrain is slightly elevated above the sea level (0-13 m), and in the hinterland it becomes a hill with dense, high and almost pathless vegetation. There is no fresh-water sources or reservoirs at Arza or in the immediate vicinity, and all the rainfalls and water solutions go to the sea. Cape Arza is a touring area, without any kind of agriculture, food production or cattle grazing.

During the first visit to the Cape we concluded that, because of the time elapsed since the NATO attacks (20 months) and the rainfalls during that period of time (over 1000 liter/m$^2$), it is almost impossible to recognize visually the places where uranium bullets struck the ground, and determine in such a way the directions of aircraft attacks, places with concentration of their fire, and outer borders of the contaminated zone. Therefore, one member of the team, a military officer who inspected Cape Arza immediately after it was targeted, was assigned to be (from time to time) a free dosimetrist who will act on his own, searching for DU penetrators and contaminated places trough vegetation randomly, guided only by his military and dosimetric experience. This proved to be very useful, because his findings enabled us to plan more rationally all our necessary activities for a successfull survey and decontamination.
We divided the area of the Cape into sections, to be able to have detailed and exact evidence and control of the work performed. The sections were rectangular, 10 m wide and 10 to 80 m long. Because the Cape was covered with vegetation which hindered a detailed dosimetric survey, the team was divided into three units: one for cutting vegetation and removing it aside on the deposition locations, one for contamination searching (dosimetry unit), and one for decontamination. All trees existing on the Cape are preserved, and only their low branches were cut.

In order to eliminate the possibility that part of a section could be overlooked by chance or less systematically checked during the survey, the section is divided by ropes into transversal paths, each one 10 m long and 0.7 m wide. Members of the dosimetry unit (2 to 6, depending on the requirements of the whole operation at a given moment) surveyed first the path visually, and after that systematically with KOMO-TL monitors. The dosimetrists walked in lines, one beside the other, with detectors which were held up to 5 cm above the ground and moved with a speed of (5 – 7) cm/s, of course much slower over the points with DU contamination and particularly at the points where the instrument count rate was slightly above the background value. The average speed of a dosimetrist along the path was about 20 m/h.

The zone of cut vegetation and systematic survey was expanded until the area with detected DU contamination was surrounded with a 30 m wide belt in which no DU contamination was found, including ricocheted penetrators and fragments.

Outside of this zone the survey was also performed. On the beach at Cape Arza a detailed dosimetric search along paths 0.7 m wide was done, but DU contamination was not discovered. On the rocky shore we did a visual and, where possible, sporadic search with instruments and a few DU penetrators were found lying on or struck in the rocks below or near the high-water mark. The rest of the Cape was also surveyed by contamination monitors as systematically as possible without clearing of bushes.

In total, the systematic contamination search was spread over an area of 37,000 m², which represents the most attractive part of Cape Arza.

The KOMO-TL instrument registers natural background radiation at Arza as (2 – 4) cps. At each place where the instrument gives a count rate higher than 4 cps, a dosimetrist puts a flag, and the decontamination unit checks later on whether this is only a background variation or really contamination by DU.

The investigation of these places was performed by the TOL/F βγ-monitor – the most sensitive instrument at our disposal. The background count rate at Cape Arza on this instrument spans a relatively large range (15 – 25) cps, depending on whether the instrument is above stones, brown or humus soil, or above terra rossa.

While above other types of ground the count rate of (20 – 25) cps was a sure indicator of DU contamination presence at the place, above terra rossa soil it was result of a relatively high content of natural uranium in the soil, which can be up to 165 Bq/kg (Vukotich et al., 1997). Not relying only on a visual identification of the type of soil in a given case, we always did a further investigation removing slowly layer by layer of soil in that place and measuring the count rate above the hole by TOL/F. If there is a penetrator or a piece of it hidden in the ground then the count rate increases with depth of the hole, and if the ground is non-contaminated terra rossa the count rate remains unchanged (holes were dug to a depth of 30 cm).
The presence of penetrator or contamination by DU dust on the ground is easy to detect and locate precisely, because the TOL/F gives in such a case high count rate accompanied with audible signal.

If a penetrator is buried in the ground, the instrument KOMO-TM is the best choice for precise localization of the penetrator pathway through the ground. Due to the specific structure of the terrain at Cape Arza (lot of stones and rocks below the surface), the penetrator after hitting the surface and entering the ground, usually already at a depth of a few centimeters, strikes a piece of stone and crushes it. Such stone turns to a gray color from uranium dust and becomes quite radioactive. If the penetrator on the way inside ground hits a large stone or a rock, it changes direction or penetrates through cracks, leaving behind a dark-gray track of uranium dust cover, which is usually easy to follow with the radiation detector. However, sometimes a lot of persistence and experience gained in the field with the instruments KOMO-TM and TOL/F was needed to find out where the penetrator disappeared after hitting a rock or between two rocks. Sometimes, the trace of DU dust showed that the penetrator slid on the rock inside ground in such way that it was expelled out of the ground again. But thanks to the dark-gray traces of uranium dust on the stones and rocks inside ground, we were able to trace buried penetrators or their fragments even up to a depth of 90 cm.

In total, we have found 35 DU penetrators on the ground surface and 139 buried in the ground, usually at a depth of 25 to 40 cm.

We have noticed that penetrators found on the ground surface were less corroded than those buried for about two years in the ground of Cape Arza. Furthermore, penetrators buried in the rocky surroundings were less corroded than those buried in the soil. Especially heavily corroded were the penetrators found shallow in the humus soil, and they almost totally disintegrate at a touch.

Around an impact point of uranium bullet, contamination on the ground surface is usually located within an area of 30 to 50 cm in radius. Digging and searching for penetrator hidden in the ground, we collected and disposed of safely all contaminated material, like soil, crushed pieces of rocks and stones, and uranium oxides around corroded penetrator. The soil very close to corroded penetrators had an activity up to $3.5 \times 10^6$ Bq/kg. In case when we found a dark-gray trace of uranium dust on a large rock inside ground, we washed it with brush and water and then collected and removed residual radioactive mud. We considered decontamination of such a place finished when we reached TOL/F count rate inside the hole less than 50 cps, i.e. a radiation level less than double value of the maximum local natural background. Finally, we refilled the hole with non-contaminated soil and stones.

Places on the ground with surface contamination only, caused by spreading out the minute parts and dust of DU penetrator after its collision with a neighboring rock, can have up to a few square meters in area. In this case we had to collect carefully dead leaves and needles, moss and thin layer of soil to decontaminate the place.

As to the mutual position of the uranium penetrators and their Al-jackets, we have not noticed any regularity. They are somewhere one beside the other, and somewhere one without the other, what is the consequence of their different penetrability and the various structures of the ground. The jackets were found, whole or in pieces, usually at a shallow depth, up to 20 cm. However, at one contamination place, where the soil was without stones in it, we found the jacket at a depth of 50 cm.
Up to now, an area of about 37,000 m² has been surveyed at Cape Arza, and DU contamination found on 16,000 m². In total 326 contaminated places on the ground were found and cleaned up. They are decontaminated below the level twice higher than the maximum value of the local natural background, which is 0.15 μSv/h (Vukotich et al., 1997). In the course of decontamination, 174 whole DU penetrators were found and removed, as well as 41 their larger fragments, which amounts to 60 kg of uranium in total. A lot of partly contaminated aluminium jackets were also found. Approximately 150 kg of highly contaminated soil (1.5·10³ - 3.5·10⁶) Bq/kg and 4 tons of the low-radioactive material (soil, pieces of rocks, dead leaves and needles etc.) were also removed.

4. CONCLUSIONS

The instruments KOMO-TL and KOMO-TM, with pancake Geiger-Müller tube, have proven high performances in searching for DU contamination. However, the TOL/F with βγ-probe, a proportional counter with large effective area and high sensitivity, has been key instrument in verifying penetrator presence in the ground and in checking quality of the decontamination performed.

Our capacities to find DU penetrators hidden in the ground were even several times better than we could expect on basis of the previous laboratory experiments. Thanks to traces of DU dust that penetrators leave behind them on the stones on their way in the ground, we were able to find them up to a depth of 90 cm.

Penetrators lying on the ground surface were less corroded than those buried two years in the ground, and penetrators buried in the rocky surroundings were less corroded than those buried in the soil. Especially heavily corroded were the penetrators found in humus soil, and they almost totally disintegrate at a touch.

Around an impact point of DU round, contamination on the ground surface is usually located within a radius of 30 to 50 cm. The soil very close to corroded penetrators had an activity up to 3.5·10⁶ Bq/kg, that is 20000 times higher than the maximum natural uranium content at the Cape. Places on the ground with surface contamination only, caused by spreading of small parts and dust of DU penetrator collided with hard objects somewhere nearby, can be up to a few square meters in area.

Collecting carefully and taking away all contaminated material (soil, pieces of rocks and stones, uranium dust and small parts of corroded penetrator, etc.) we have decontaminated the site below the level twice higher than the maximum value of the local natural background.

REFERENCES


