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On problems related to the deployment of depleted uranium weapons in the Balkans

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The likely long-term environmental and health effects of the deployment of weapons containing depleted uranium (DU) in the Balkans are discussed. To determine whether depleted uranium or spent reactor fuel was used in the weapons, knowledge is required of the ^{235}U to ^{238}U activity (or concentration) ratio in the measured samples. To this end, and to distinguish between uranium originating from natural and man-made sources, we discuss some of the methodology and metrology issues involved in performing alpha- and gamma-spectrometry of uranium in environmental and human samples. We present results of nuclear spectrometry performed on DU core deposits eluted from the aluminium jacket of a PGU-14 bullet found in South Serbia. We draw attention to aspects involving ionising radiation, which are likely to be of importance when formulating a prognosis of the possible environmental and health impact of the deployment of DU weapons, indicating the importance of the inhalation pathway in children.

KEY WORDS: *Uranium; Nuclear Warfare; Radioactive Pollutants; Health; Environmental Pollution; Yugoslavia; Europe*

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INTRODUCTION

Deployment of weapons containing depleted uranium (DU) in the Middle East during the Gulf War (in 1990 and 1991) and later in the Balkans: in the Republic of Srpska (1995), Kosovo, South Serbia and Montenegro (in 1999), raises the question of long-term environmental and health effects of the ionising radiation from such weapons. Natural uranium is commonly present in the environment and some uranium is always found in human urine. As some of the regions in the Balkans where DU weapons were used are also regions of enhanced natural background radiation (due mainly to the uranium and thorium decay series), it is important to distinguish between the different sources of radiation burden to the population involved. Therefore, very careful isotopic composition studies should be made to distinguish between natural and depleted uranium in the evaluated urine samples. It is also interesting to know whether depleted uranium or spent reactor fuel was used in the weapons. Exact knowledge on the ^{235}U to ^{238}U activity (or concentration) ratio in the material used in uranium weapons is therefore of particular interest. We discuss some of the methodology and metrology issues involved in performing alpha- and gamma-spectrometry of uranium. We present results of nuclear spectrometry performed on DU core residues taken from the aluminium jacket of a PGU-14 bullet found in South Serbia. We also review the radiation hazard due to uranium weapons, taking into account the difference between climatic and environmental conditions of the Balkans and the Middle East. We wish to draw attention to those aspects which are likely to be of importance when formulating a prognosis of the environmental impact of the deployment of depleted uranium weapons in any future conflicts in Europe.

THE DEPLETED URANIUM WEAPON

The concept of using uranium cores in anti-armour ammunition was considered as early as the end of the Second World War. Depleted uranium, a waste product of the nuclear industry, is now used in weapons produced by many countries all over the world. The most widely used cannon which fires ammunition containing depleted uranium (DU) is the General Electric GAU-8/A "Avenger". This is a 30 mm, seven-barrel Gatling gun (1), mounted on board of a strike and tank-buster aircraft, the Fairchild-Republic A 10A "Thunderbolt II" (also known as the "Warthog"). The aircraft was designed in 1970, mainly for fighting Warsaw Pact tanks and troop carriers in a potential total conflict in Europe. As it had been envisaged that in such a conflict nuclear weapons would also be

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used, the environmental impact of the use of depleted uranium was not considered in any great detail.
The GAU-8/A "Avenger" gun uses 1174 rounds. Within the so-called "Combat Mix Ammunition", 20% of this number are filled with high explosive (PGU-13), and 80% use a DU core (PGU-14). A total number of 148 A-10 aircraft were deployed in Saudi Arabia firing 783,514 rounds, which resulted in about 260 tons of DU being deposited during the Gulf War (1). About 30,000-40,000 of DU rounds were used in the Balkans during the 1999 conflict.

SOME PHYSICO-CHEMICAL PROPERTIES OF THE PGU-14 WEAPON

The DU core of a PGU-14 bullet, often called a DU penetrator, has the form of a cylinder of 15-mm diameter and 70 mm length, and a truncated conical top. This core is housed in an aluminium jacket. The mass of the DU core of a PGU-14 bullet is about 300 g, the cylindrical part of it weighing about 240 g, which is almost exactly the mass of 1 M of ^{238}U. One mol is the typical amount of ^{238}U in 100 tons of soil. The activity of one mol of ^{238}U is:

$$A = \lambda N = \frac{\ln 2}{4.5 \cdot 10^9 \cdot 365 \cdot 86400} \cdot 6.02 \cdot 10^{23} s^{-1} = 2.94 MBq$$

For future rough estimates we can use the value of 3 MBq for the alpha-activity of 238U in a single bullet. Uranium is in equilibrium with its short-lived daughters, ^{234}Th ($T_{1/2}$=24.1 days) and ^{234m}Pa ($T_{1/2}$=1.18 min), which contributes another 6 MBq of beta-activity. The daughters also emit gamma radiation. The depletion is never completed, meaning that another 0.6 MBq arises from ^{234}U, and only 30 kBq from ^{235}U. The daughters of these uranium isotopes are longer-lived and consequently of lower specific mass activity. Therefore their activity is not considered here. The initial velocity of the bullet is V=1060 m/s (≈3.2 M). This allows one to estimate the kinetic energy of the core:

$$E_{kin} = 0.5 mV^2 \approx 119\ kJ/mol \quad (2)$$

Let us assume that as the bullet strikes the tank armour, an adiabatic process occurs (i.e. that there is no loss of heat). Then, the kinetic energy of the bullet is wholly converted to heating up the uranium core and to melting the tank's armour, in a cylinder along the core's path, hence, from first principles:

$$E_{kin} = (m_u c_u + m_a c_a)\Delta t \quad (3)$$

where: m_u (kg) is the uranium core mass (1 mol), m_a is the mass of the heated fragment of armour, and c_u and c_a (J K^{-1} kg^{-1}) are the specific heat of uranium and armour material (steel), respectively, while Δt (K) is the increase of temperature.
The specific heat of uranium is c_u=117.6 J/(K kg). For a rough estimate, one can assume that c_u=c_a, m_u=m_a (corresponding to steel armour of about 20 cm thickness):

$$\Delta t \approx \frac{E_{kin}}{2 m_u c_u} \approx 2100 K \quad (4)$$

This temperature exceeds the melting point for steel and uranium, showing why the uranium core can penetrate the tank's armour. However, far more energy is in fact released as the melted uranium encounters oxygen from the air inside the attacked target, according to the formula:

$$U + O \rightarrow UO + 759\ kJ/mol \quad (5)$$

Thus, heat released from oxidation of the uranium core inside the target can exceed the kinetic energy of the bullet by a factor of up to 6.5.

ON THE METHODOLOGY AND METROLOGY OF NUCLEAR SPECTROMETRY OF URANIUM

The following comments on methodology and metrology should facilitate the interpretation of nuclear spectrometry of uranium samples. Nuclear gamma-spectrometry determines the ratio of the gamma activities of ^{235}U and ^{238}U and not the ratio of masses of these isotopes. For natural uranium, the mass ratio ^{235}U/^{238}U should be about 0.7%, as is well known. However, specific activities of these isotopes are different: ^{235}U is more active than ^{238}U, the half-life of ^{235}U being 7.0510^8 years while that for ^{238}U is 4.4710^9 years. The ratio between the reciprocals of these two half-life values is the ratio of ^{235}U/^{238}U activity, and it is 6.34, if the masses of both isotopes are equal. In other words, a given mass of ^{235}U shows an activity 6.34 times higher than the same mass of ^{238}U would show. In natural uranium, only 0.7% of the mass is ^{235}U, but it is 6.34 more active, so in the end: 0.7 %x 6.34 = 4.5% of the total activity is due to ^{235}U.
As far as metrology is concerned, the activity of ^{238}U can be determined either by alpha-spectrometry or by gamma-spectrometry, through ^{234m}Pa (or ^{234}Th) peaks. After about 150 days, ^{238}U and ^{234}Th and ^{234m}Pa arrive at an equilibrium, which means that the ^{234m}Pa and ^{234}Th activity is equal to the ^{238}U activity. The activity of ^{235}U cannot usually be seen directly in the alpha spectrum, so in practice it is only the 185.7 keV gamma line of ^{235}U that is used. There may be two problems connected with this:
1. A ^{226}Ra gamma-line of almost exactly the same energy exists, and ^{226}Ra is present in small amounts everywhere. This, howev-

er, should be of no concern in the case of measurements of DU.
2. The penetration of the 185.7 keV gamma-ray line of ^{235}U in metallic uranium is very small, the mean free path being only 0.4 mm (the density of pure metallic uranium is huge: 19 000 kg/m^3, which is the main reason why the military uses it in the first place!). Due to this self-shielding, the gamma-detector in the spectrometer measures only a very small fraction of the sample. On the other hand, the gamma-ray line used for the determination of ^{238}U has a much higher energy, namely 1001 keV, so it is much less shielded. The difference in shielding between the two lines can seriously distort the ratio of the measured activities, causing an underestimation of the ^{235}U activity. Since in the process of producing nuclear fuel, natural uranium is depleted to about 20% or 30% of its original ^{235}U content, a depletion ratio of about 25%, with respect to the natural ^{235}U-content, seems reasonable. An even larger depletion of ^{234}U may also be normal, since it is a lighter element than ^{235}U, the depletion in isotope 234 is larger and ^{234}U has a very long half-life time (245000 years), so it cannot re-grow from ^{238}U. It is well known that the studies on isotopic ratios in environmental samples can provide data on the origin of many man-made radionuclides. This may also be the case for uranium used in DU weapons. Three uranium isotopes: ^{238}U, ^{235}U and ^{234}U, are present in the environment. Although ^{238}U and ^{234}U are members of the same uranium series, their ratio in environmental samples is not very well defined. For mineral samples it is usually below 1, for water samples it usually exceeds 1 (2). The reason for this is in the ability of water to leach out the uranium atoms from a mineral grain prior to their alpha-decay and after its decay, when it has recoiled from its original place in the mineral lattice, the local structure being destroyed by the alpha-particle. The natural ^{235}U to ^{238}U ratio is very well defined, therefore it appears to be more suitable for studying the origin of uranium. However, the activity of ^{235}U is very low, so its determination is always less certain than that of ^{234}U. Let us consider the case when the activity of ^{235}U can be measured accurately enough. One can then distinguish between the natural components of uranium in a DU sample by solving the following set of equations:

$$\begin{aligned} A_{238} &= A_n + A_d \\ A_{235} &= \zeta A_n + \xi A_d \end{aligned} \qquad (6)$$

where:

A_{238} or A_{235} are the observed specific activities of ^{238}U or ^{235}U, respectively, A_n is the *a priori* unknown natural component of ^{238}U activity, A_d is A_n, but for the weapon (depleted) component, ζ is the natural ^{235}U-to-^{238}U activity ratio, equal to 0.045, and ξ is the ^{235}U-to- ^{238}U activity ratio in the DU weapons (expected to be about 0.01).

The solutions of the above set of equations (equ. 6) are:

$$A_d = (\zeta A_{238} - A_{235}) / (\zeta - \xi) \qquad (7)$$

$$A_n = (A_{235} - \xi A_{238}) / (\zeta - \xi) \qquad (8)$$

Thus, the DU fraction, F, defined as:

$$F = (A_d/A_{238}) \qquad (9)$$

can be calculated, in percent, from the formula:

$$F = \{[\zeta - (A_{235}/A_{238})] / (\zeta - \xi)\} \cdot 100\% \qquad (10)$$

For samples with a well-defined natural ratio of ^{234}U-to-^{238}U, formulae similar to those above can be applied to identify the origin of uranium in the sample.

Either gamma- or alpha-spectrometry can be used to determine the ratio of ^{235}U-to-^{238}U activities. The method applied should depend on the type of sample. Evaluation of this ratio using gamma-spectrometry in fragments of bullets would appear to be more appropriate for determining the origin of DU used in the weapons. Since the sensitivity of alpha-spectrometry is far better (at the expense of a much more laborious sample preparation technique), it would be more advantageous in assaying biological, medical and environmental samples. If available, mass spectrometry (ICP-MS) is, of course, another very powerful and accurate technique.

NUCLEAR SPECTROMETRY OF A PGU-14 BULLET CORE

Depleted uranium residues extracted from an aluminium jacket of a PGU-14 bullet (30 mm diameter), fired from a GAU-8 "Avenger" anti-tank gun of a Fairchild-Republic A10-A airline, found in South Serbia (Bratoselce field location), was analysed at the Institute of Nuclear Physics in Kraków, using gamma- and alpha-spectrometry. The aim was to determine the uranium depletion rate and to find possible traces of accompanying nuclides. Gamma spectrometry was performed on the complete aluminium jacket of the bullet core (the core itself was not available for measurement, so only the core residues remaining on the jacket were evaluated). For alpha-spectrometry, the jacket was placed in a 0.01 M HCl solution and boiled for 20 minutes. Next, the solution was filtered, the residue evaporated to dryness and the dry residue re-dissolved in 9M HCl. This solution was then passed through an anion exchange column filled with Dowex-1. Uranium was retained on the column. It was next eluted with nitric acid and, finally, a NdF_3 co-precipitated alpha-spectrometry source was obtained.

Only the expected nuclides, namely ^{235}U, ^{234}U, ^{238}U, ^{234m}Pa and

^{234}Th, were observed in the spectra (see Figure 1). Both spectrometry methods gave similar results. The depletion rate was not high, the ^{235}U / ^{238}U activity ratio being about 0.01, confirming that about 20÷25% of the ^{235}U natural abundance was still present in the uranium core of the bullet, this being representative of depleted uranium. Comparison of the obtained results using gamma- and alpha- spectrometry suggests that gamma-spectrometry is perhaps more useful for studies of the U isotopic ratio and its variety in bullets, since the obtained precision is similar, alpha-spectrometry requiring more laboratory work. From nuclear spectrometry analysis of the bullet jacket, it seems that the bullet consisted of depleted uranium originating from fuel production rather than from reprocessed spent fuel, as in the latter case also ^{236}U and probably traces of plutonium should have been observed.

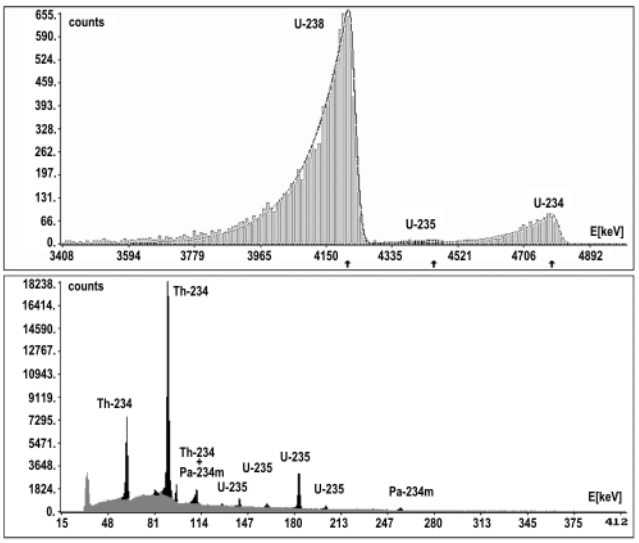

Figure 1. Selected parts of alpha- (above) and gamma-ray (below) spectra obtained from an aluminium jacket of a PGU-14 bullet with a depleted uranium core deployed in South Serbia (Bratoselce field location)

ON THE HEALTH IMPACT OF DEPLETED URANIUM

While uranium is not an abundant natural element, it is distributed quite widely. The typical total content of U within the human body is 90 μg (3), giving a total activity of 1.1 Bq of ^{238}U (1 μg of U yields 12 mBq of ^{238}U). The main sites of uranium deposition in the body are the skeleton (59 μg) and the kidneys (7 μg). The daily intake of U is 1.9 μg. It is removed from the kidneys with a biological half-life time of 6 days, whereas for the skeleton two components have been distinguished: a fast one, of 20 days and a slow one, of 5000 days.

About 750 000 PGU-14 bullets fired during the Gulf War contained a ^{238}U activity of about 1.27 TBq (plus twice that, from ^{234m}Pa and ^{234}Th). Military reports gave the number of 4132 targets destroyed, which, on the average, gives 190 bullets fired per target (an average series lasting 2.7 s). Therefore, a rough estimate of the activity released at any one target yields 570 MBq ^{238}U + 570 MBq ^{234}Th +570 MBq ^{234m}Pa + c.a. 100 MBq ^{234}U and 5.7 MBq ^{235}U. A similar deposition pattern may be assumed for an average target site in the Balkans. However, the total activity deposited during the Balkans intervention was 30-40 times lower than in the Gulf War, since 30-40 times fewer bullets were deployed.

A (most likely, conservative) approximation, assuming that this activity is deposited on the surface of the earth over a rectangular area around the target, of size 10 m x 100 m = 1000 m^2, results in approximately: 570 kBq/m^2 of ^{238}U (plus the same activities of ^{234m}Pa and ^{234}Th), plus 100 kBq/m^2 of ^{234}U, and plus 5.7 kBq/m^2 of ^{235}U. The external exposure due to this deposited activity will follow mainly from ^{234m}Pa (4). The surface activity to dose equivalent rate conversion factor for this nuclide is $3.30 \cdot 10^{-5}$ [(Sv/year)/(Bq/cm^2)], and for ^{234}Th it is an order of magnitude lower. The ambient equivalent dose rate from such a deposit on the earth surface can be thus estimated to be about 230 nSv/h, some 4 or 5 times higher than that from natural sources. However, in the 1999 Balkan conflict most DU cores appear to have penetrated some depth into the ground, hence their radiation at the surface would have been strongly attenuated by the soil. The given values are therefore upper estimates and, for practical purposes, potential external gamma-ray exposure of the population from DU weapons deposited on the ground, is not expected to significantly contribute to that from natural background.

When assessing other potential sources of radiation hazard to the population, it appears that of major importance are exposures due to the inhalation pathway, as suggested by the activity-to-dose conversion factors published in (3), presented in Table 1. According to this table, new-born children, after inhaling AMAD=1μm aerosols containing ^{238}U oxide of activity 40 Bq, will incur an individual dose equivalent of 1 mSv, while adults will incur the same dose after inhaling an activity of 125 Bq under the same conditions. This translates to inhaling a little over 3 mg or about 10 mg of ^{238}U (in the form of oxides) by the newborn child and adult, respectively. For the ingestion pathway, an individual dose equivalent of 1 mSv will be incurred after ingestion of about 3 kBq or of 22 kBq of ^{238}U by a newborn child or an adult, respectively.

Table 1. Dose conversion factors (ICRP 30, 1979) for ^{238}U [Sv/Bq]

Dose conversion factors (ICRP, 1979) for ^{238}U [Sv/Bq].

	New-born children	Adults
h_g (inhalation, AMAD = 1 μm)	$2.5 \cdot 10^{-5}$	$8.0 \cdot 10^{-6}$
h_g (ingestion)	$3.4 \cdot 10^{-7}$	$4.5 \cdot 10^{-8}$

(AMAD-Aerosol Mean Aerodynamic Diameter)

CONCLUSION

Gamma- and alpha- nuclear spectrometry techniques are able to resolve between the natural and man-made sources of uranium contributing to the health and environmental impact of the deployment of DU weapons. It is also possible, in principle, to distinguish the presence of burnt-out nuclear waste in the DU used in the weapons. Alpha-spectrometry is more appropriate for assaying the uranium content in human and biological samples. When considering the radiation hazard to the population from the deployment of DU weapons, external gamma-irradiation is relatively unimportant, unlike the inhalation pathway, especially in children. Among the civilian general population inhabiting the areas stricken by DU weapons, the critical group may consist of children who by playing inside wrecked military targets (tanks) could inhale uranium dust present there in large quantities. Ingestion of natural and depleted uranium by the civilian population should be analysed. Assay of urine samples for uranium in different cohorts in the population seems to be the most prospective form of assessing the body burden from either pathway. However, due to the presence of areas of naturally enhanced background radiation in the studied Balkan regions, the selected assay method should enable one to distinguish between natural and depleted uranium. Additionally, suitably selected environmental samples should be assessed in order aid the interpretation of results of deployment of DU weapons in regions of normal and high natural background.

The subject of the pharmaco-toxic pathways of uranium in the human body, possibly additionally involving the radiation impact of uranium decaying in selected organs of the body (such as kidneys or lymph nodes), is a matter of separate consideration and has not been studied here.

The climatic and environmental conditions in the Balkans and in the Middle East are different. The Kosovo area seems to be of special interest, as it is a densely populated area with a moderate climate and a variety of natural background levels. The obtained data may be important not only for the population of Kosovo, but also for developing a prognosis of the most likely health hazard pathways, resulting from any future deployment of uranium weapons in countries of a similar climate and population pattern.

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REFERENCES

1. Pike J. GAU-8 Avenger, 2000. Available from: http://www.fas.org/man/dod-101/sys/as/equip/gau-8.htm
2. Asikainen M. State of disequilibrium between ^{238}U, ^{234}U, ^{226}Ra, ^{222}Rn in groundwater from bedrock. Geochimica et Comologica Acta 1981;45:201.
3. ICRP 1979, ICRP Publication 30 with Supplements. Pergamon Press; 1979.
4. Kocher DC. Dose-Rate Conversion Factors for External Exposure to Photons and Electrons. Health Physics 1983;45(3):665-86.