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CHARACTERIZATION OF DEPLETED URANIUM AND ITS DANGER FOR INHALATION

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1. Introduction

The following technical note make the assumption that the reader has the suitable scientific background contained in Ref. 1[1]. This note makes the specific analysis regarding the specific problem of the characterization of Depleted Uranium (DU) considering the presence of Plutonium. This note will consider these basic assumptions:

- The DU is a mixture of radio nuclides (U238, U235,, U234) with some well known concentrations.
- There have been during the last months and particularly in the last month of January, contradicting news about the dangers arising from DU for the presence in it of Plutonium.
- If DU comes out from the cycle of enrichment of uranium, it is not possible to find in it U236 nor Pu239, unless only "fresh" uranium is used, that is uranium which is freshly extracted from the mines or coming from processing of natural uranium. If, on the contrary, DU arises from an enrichment progress which employs as rough material also uranium coming from the reprocessing cycle of spent nuclear fuel, then all these and other nuclides can be present in DU.
- We will call the first one as "**Clean DU**", and the second as "**Dirty DU**", without, by this, considering the clean DU as a safe material.
- The fact that DU used in the Balkan area is, at least partly, "dirty" has been confirmed by UNEP (United Nations Environmental Program) which has verified[2] the presence of U236 in DU.

[1] M. Cristaldi, A. Di Fazio, C. Pona, A. Tarozzi, M. Zucchetti, "Alcune tesi e fatti sull'uranio impoverito (DU), sul suo uso nei Balcani, sulle conseguenze sulla salute di militari e popolazione", (in corso di pubblicazione su "Giano" n.36), vedi anche il sito: http://www.peacelink.it/tematiche/disarmo/u238/documenti/uranio_impoverito.html

[2] http://balkans.unep.ch/press/press010116.html

The necessary starting point will be a correct information about composition of DU. Far beyond the mixture of the 3 major nuclides, we should know, in the case of dirty DU from which particular nuclear plants originates the uranium to "reprocess" and its characteristics. Eventually, even if we know exactly the original composition of clean DU and dirty DU, we should know also in which amount they have been mixed to produce DU ammunitions.

We will refer to a document published by WISE Uranium Project Fact Sheet[3] (WISE), which exhibits some calculations based on the data given by the U.S. Dept. of Energy, and verified by one of the authors (M.Z.) as right.

We will hence refer to these news:

- The dirty DU used comes from the enrichment plant of Paducah (source: DOE).
- The compositions of clean DU and dirty DU are those reported in the cited document, excluded the presence of some decay products not cited, for which, as we will see later, we will make some assumptions.
- The presence of some other fission products in dirty DU (as Cs137, Sr90 and so on) although it cannot be excluded "a priori", it is not considered, because these elements are very easily separated from uranium.

It will be obvious be possible to discuss these assumptions, but we will refer to them for simplicity, and to illustrate the method with which we can characterize DU and its hazardousness.
We will make hence a comparison between clean and dirty DU following these assumptions.

2. Decay products of Uranium: the question of the "missing daughters".

Every radioactive nuclide (or radionuclide) decades in an other nuc lide, which cam be radioactive on its own or stable. These decay products, when are radioactive, contribute, when present to the total radioactivity of the material where the "father" radionuclides are.

- In this way, U238 brings with itself their decay product Th234, which decays in Pa234m, which decays in U234 and so on (see below for the following of the chain).
- Similarly, U235 generates Th235, which generates Pa231, which generates Ac227, which decays in Th227, which decays in Ra223, which again generates Rn219. But Rn219, as it is a noble gas, is removed from the solid matter, and the chain stops here.

[3] http://www.peacelink.it/tematiche/disarmo/u238/documenti/durepe.pdf

- U234, eventually, produces Th230, which decays in Ra226, which decays in Rn222, which is gaseous and escapes from the solid and the chain stops here.

Hence in the clean DU, in addition to U238, U235 and U234, we must take into account for the presence of the whole radioactive chain "not gaseous" consisting of Th234, Pa234m, Th231, Pa231, Ac227, Th227, Ra223, Th230, Ra 226.

One should ask what would be the concentration of these "daughter" nuclides. Luckily there is an easy answer: at equilibrium its radioactivity will be the same of the "father" nuclide.
For example, 1000 Bq/g of U238 means also 1000 Bq/g of Th234, 1000 Bq/g of Pa234m and so on.
As uranium is present in nature since ever (billions of years), all its daughter nuclides are present at equilibrium.
Let's have a look now to the table for clean DU in the WISE document, and we will see that some of these nuclides are missing.

Regarding the dirty DU, we will consider the table in the WISE document at which we will must nevertheless add the following:

- U236 decays in Th232, from which generates a radioactive chain which is non relevant for our purposes, as we will se later.
- Np237 decays in Pa233, and this in U233, from which another non relevant chain originates.
- Pu239 decays in U235 and from here with the already mentioned chain.

If now we have a look to the table relative to dirty DU in the WISE document, we will note that here also some nuclides are missing.

On the other hand, the missing nuclides are characterized by very long half life (more than 1000 years) or which in the chain are generated by such a nuclide. Nuclides with very long half lives reach the equilibrium after a lap of time comparable with their half life.

During the enrichment (or depletetion) process of uranium, it is theoretically separated from all other chemical species and hence also from its radioactive descendants, even some loss of efficiency in the process may leave some of these isotopes). Uranium would become again "new", as just created, and it would start again from zero, to create its daughters. Those a long half life (for example Th230) do not reach the activity of the father in the few decades of life of this new

uranium (before it is employed somewhere), and hence their concentration at the moment of utilization should be very little and negligible.
Other nuclides, as all the long lived daughters of U236, Np237 and Pu239 (for example Th232 and U233), are originating from a "new" nuclide (born during the utilization as fuel in the reactor), and hence it is certain that their presence is negligible in dirty DU, when it becomes a DU bullet.

On the other hand, we must also take into account that:

- The chemistry of some actinides such as thorium is very similar to that of uranium, and hence we cannot say that thorium is totally absent in "new" uranium. Nevertheless, if we consider that the separation between the various isotopes of uranium is base on physical properties (such as weight) instead of chemical properties, it is more likely that thorium is found in the enriched fraction.
- DU, after fired, "in situ" (in the environment of Balkans or in the body) starts generating daughter radioactive isotopes, but many of them are long lived radioisotopes and hence they do not represent a problem for human beings.
- The last contribution will become relevant after centuries and thousands of years and it is non considered here.

Summarizing, we can look at tables 1a and 1b, 2a and 2b. Tables 1a and 1b characterize an "old" DU, either "clean" and "dirty", both considered at equilibrium with their descendants, included the long lived, non gaseous, ones, generated by the primordial isotopes such as U238, U235 and U234.

Tables 2a and 2b characterize the "new" DU, either "clean" and "dirty", both at equilibrium with their non gaseous decay short lived products. We will consider the latter for our dose calculations.

The radioactive decay schemes of the isotopes considered may be seen at the URL http://www.dne.bnl.gov/CoN/index.html.

Table 1a: characterization of clean DU. Specific activity in Bq/g. Initial concentration: U238 (99.8%), U235 (0.2%), U234 (0.000821%). All the non gaseous nuclides have reached the equilibrium ("old" uranium), except U234 generated by U238, for which we assume the activity shown in the document of the WISE.

Nuclide	**Specific activity (Bq/g)**	**Note**
U238	1.243e+4	Nuclide father
Th234	1.243e+4	At equilibrium with U238
Pa234m	1.243e+4	At equilibrium with U238
U235	1.6e+2	Nuclide father

Th231	1.6e+2	At equilibrium with U235
Pa231	1.6e+2	At equilibrium with U235
Ac227	1.6e+2	At equilibrium with U235
Th227	1.6e+2	At equilibrium with U235
Ra223	1.6e+2	At equilibrium with U235
U234	1.9e+3	Nuclide father
Th230	1.9e+3	At equilibrium with U234
Ra226	1.9e+3	At equilibrium with U234
(TOTAL)	**4.4e+4**	

Note that: 1.243e+4 = $1.243\ 10^4$

Table 1b: characterization of dirty DU. Specific activity in Bq/g. Initial concentrations: U238 (99.57%), U235 (0.2%), U234 (0.001939%), U236 (0.22%), Pu239 (4.401e-07%), Np237 (2.469e-05%). All the non gaseous nuclides have reached the equilibrium ("old" uranium), except U234 generated by U238, for which we assume the activity shown in the document of the WISE.

Nuclide	**Specific activity (Bq/g)**	**Note**
U238	1.240e+4	**Nuclide father**
Th234	1.240e+4	At equilibrium with U238
Pa234m	1.240e+4	At equilibrium with U238
U235	1.6e+2	**Nuclide father**
Th231	1.6e+2	At equilibrium with U235
Pa231	1.6e+2	At equilibrium with U235
Ac227	1.6e+2	At equilibrium with U235
Th227	1.6e+2	At equilibrium with U235
Ra223	1.6e+2	At equilibrium with U235
U234	4.48e+3	**Nuclide father**
Th230	4.48e+3	At equilibrium with U234
Ra226	4.48e+3	At equilibrium with U234
U236	5.43e+3	**Nuclide father**
Np237	6.44e+0	**Nuclide father**
Pu239	1.01e+1	**Nuclide father**
(TOTALE)	5.7e+4	

Note that: 1.240e+4 = $1.243\ 10^4$

Table 2a: characterization of clean DU. Specific activity in Bq/g. Initial concentration: U238 (99.8%), U235 (0.2%), U234 (0.000821%). Only the short lived non gaseous nuclides have reached the equilibrium.

Nuclide	**Specific activity (Bq/g)**	**Note**
U238	1.243e+4	Nuclide father
Th234	1.243e+4	At equilibrium with U238
Pa234m	1.243e+4	At equilibrium with U238
U235	1.6e+2	Nuclide father
Th231	1.6e+2	At equilibrium with U235
U234	1.9e+3	Nuclide father
(TOTALE)	**3.95E+4**	

Si legga: 1.243e+4 = $1.243\ 10^4$

Table 2b: characterization of dirty DU. Specific activity in Bq/g. Initial concentrations: U238 (99.57%), U235 (0.2%), U234 (0.001939%), U236 (0.22%), Pu239 (4.401e-07%), Np237 (2.469e-05%). Only the short lived non gaseous nuclides have reached the equilibrium.

Nuclide	**Specific activity (Bq/g)**	**Note**
U238	1.240e+4	**Nuclide father**
Th234	1.240e+4	At equilibrium with U238
Pa234m	1.240e+4	At equilibrium with U238
U235	1.6e+2	**Nuclide father**
Th231	1.6e+2	At equilibrium with U235
U234	4.48e+3	**Nuclide father**
U236	5.43e+3	**Nuclide father**
Np237	6.44e+0	**Nuclide father**
Pa233	6.44e+0	At equilibrium with Np237
Pu239	1.01e+1	**Nuclide father**
(TOTALE)	**4.74e+4**	

Note that: $1.240e+4 = 1.240\ 10^4$

3. The real composition of dirty DU

Regarding dirty DU, we to consider also, as noted by the WISE document, that the composition is based on the assumption that all Pu239 and Np237 are not separated from uranium during the re-processment of nuclear fuel. This obviously overestimate the real amount of such as isotopes in DU. For our purposes, this is not really a problem: the higher danger of dirty DU is not related to Pu or Np, but to the higher amount of U234 and U236 (not present at all in clean DU).
Even if we assume that Pu and Np are completely separated because they represent a different chemical compound than uranium, it is not possible to separate U234 and U236, which are chemically undistinguishable from U238.
Hence, we assume that Pu and Np are present in the maximum amount. We will verify that this fact implies only a negligible contribution to the total dose (and to the global risk) of less than 1%. Their presence, is not relevant, except in one case (see later).

Plutonium may generate fears non only for its radiological toxicity, but also for its chemical toxicity. But, following the statements contained in the WISE document (that we have shown are overestimated), we can easily calculate that the amount of Pu in dirty DU is about to 4.4 mg every tonne of DU. Considering that NATO has spread between 15 and 30 tonnes of DU over the environment and people of Yugoslavia, the total amount of DU is around 100 mg or little more. This is not, absolutely, a small quality, but when it is spread over a large territory it is diluted to very low concentration. Even though the lethal dose for a human being is about 1 microgram, this

quantity is not likely to be ingested by a single person because it correspond to a mass of 225 g of DU, which is much more than the lethal dose (for chemical toxicity) for uranium.
Radio toxicity does not have, for delayed and stochastic effects to happen, any threshold: we must to calculate the risk due to an higher exposure to radiation and then to evaluate if this is relevant or negligible.

4. Dose calculations

In order to evaluate the danger for a specific nuclide, we must evaluate the Effective Dose Equivalent (EDE) to man.

Referring to the analysis already reported in this work, to obtain the values of dose, it is enough to multiply the figures already given by the amount of DU.
In this section we only wish to make a comparison between clean and dirty DU.
The base scenario will be:

1. immediate inhalation of uranium aerosol during attack (1 hour).
2. release of 1 g of DU at the level of the soil.
3. atmospheric dispersion evaluated by the Pasquill model (non-homogeneous dispersion) and most unfavourable direction (highest exposure)
4. distance: 1 km from explosion
5. human body modelled following ICRP recommendations.

The calculation of the dose to population is made using the program called GENII[4], which is a software elaborated by an US laboratory, internationally acknowledged and utilised.

5. Results, Analysis and Conclusions.

The main result is the following:

- **Dirty DU: $1.1\,10^{-7}$ Sv/g**
- **Clean DU : $6.9\,10^{-8}$ Sv/g**

[4] B.A. Napier et al. (1990), GENII – *The Hanford Environmental Radiation Dosimetry Software System*, PNL-6584, Pacific Northwest Laboratories (USA)

these values represent the EDE which is committed for life (50 years) after 1 hour of inhalation at 1 km from the site of release of DU. Obviously, when the distance is different, also the values change.

Obviously the highest committed dose is given during the first year after inhalation:

- **Dirty DU: $2.3\,10^{-8}$ Sv/g**
- **Clean DU : $1.4\,10^{-8}$ Sv/g**

It is also easy to verify that the external exposure to the radiation of DU is negligible, because these are essentially only alpha emitters.

We wish now to answers to a couple of important questions.

- Which nuclides contribute most to the dose?
- Which are the organs most exposed?

The answer to the first question is given in tables 3.1 and 3.2 (single nuclide) and 3.3 (nuclides grouped for "families"). Table 4 wishes to answer to the second question.

Table 3.1 – Clean DU. Contribution from single nuclides to the EDE. Sv/g released.

Nuclide	**Sv/g**	**%**	**Note**
U238	5.8e-8	84%	Nuclide father
U234	1.0e-8	15%	Nuclide father
U235	8.3e-10	1.2%	Nuclide father
Th234	1.7e-11		daughter of U238
Pa234	1.0e-12		daughter of U238
others	negligible		
Total	**6.9e-8**		

Table 3.2 - Dirty DU. Contribution from single nuclides to the EDE. Sv/g released.

Nuclide	**Sv/g**	**%**	**Note**
U238	5.7e-8	52%	Nuclide father
U236	2.8e-8	25%	Nuclide father
U234	2.5e-8	23%	Nuclide father
U235	8.3e-10		Nuclide father
Np237	1.7e-10		Nuclide father
Pu239	1.2e-10		Nuclide father
Th234	1.7e-11		daughter of U238
Pa234	1.0e-12		daughter of U238
others	negligible		
Total	**1.1e-7**		

Table 3.3 - Contribution to the EDE from "families". Sv/g released

Nuclide father	**Clean DU**		**Dirty DU**	
	Sv/g	**%**	**Sv/g**	**%**
U238 and daughters	5.8e-8	84%	5.7e-8	51%
U235 and daughters	8.3e-10	1.2%	8.3e-10	0.8%
U234 and daughters	1.0e-8	15%	2.5e-8	23%
U236 and daughters	--	--	2.8e-8	25%
Np237 and daughters	--	--	1.7e-10	0.2%
Pu239 and daughters	--	--	1.2e-10	0.1
TOTAL	**6.9e-8**		**1.1e-7**	

Table 4. – Dose to single organs. Comparison between clean and dirty DU, Sv/g released.

Targeted Organ	**Clean DU Sv/g**	**Dirty DU Sv/g**	**Ratio: (dirty)/(clean)**
Lungs	5.7e-7	9.3e-7	1.6
Kidneys	1.1e-10	1.8e-10	1.6
Bone Surface	3.7e-11	5.1e-9	140
Bone Marrow	3.7e-12	4.1e-10	11
Gonad	8,7e-13	4.6e-11	53
Gut	1.3e-10	3.4e-10	2.6
Other organs	---	---	
Total *	**6.9e-8**	**1.1e-7**	**1.6**

** to obtain the total EDE, multiply the dose to a single organ by the correspondent weighting factor.*

We can conclude that:

- the risk for inhalation of dirty DU is about 1.6 times the clean. The increase is almost totally given by the increase of the dose to lungs. This value can be corrected by the amount of dirty and clean DU effectively use by NATO in Yugoalsvia.

Clean DU:

- 84% of the dose arises from U238 and its daughters.
- 15% of the dose arises from U234 and its daughters. As a consequence U234 is not negligible.

Dirty DU

- the increase of the dose is highly due to the presence of U234 and U236 (48% of the total)
- 51% of the dose arises from U238 and its daughters.
- The contribution of Pu to the dose in negligible. It is not relevant for the dose calculations to know weather it has been or not completely removed during reprocessing of nuclear fuel. What is relevant is the presence of U234 and U236. only for the dose to bone marrow Pu is not negligible (see below).

- Np237 and daughter contribute only for 0.2%. we can do the same considerations as for Pu. Also in this case its contribution to bone marrow is not negligible.

Most targeted organs

- the target organs for inhalation are obviously the lungs.
- Kidneys and gut are also exposed to dose, because this is the pathway uranium walks during the excretion from human body.
- There are some other organs which are exposed, in particularly the surface of bones and the bone marrow.
- The value of dose to these organs is relatively small. This is not surprising because only a small fraction of DU is retained in bones and irradiates the bone marrow.
- Comparing the values for dirty and clean DU, we can see a very big increase of the dose to some organs, such as bones, bone marrow and gonads (relevant for the genetic consequences).
- Regarding the bone marrow, we can see that the main contribution (73%) comes from Np237 (dirty DU), which is as a consequence relevant. The contribution of Pu is 24%, which is also not negligible.
- These results, in particular the value of the dose to bone marrow, are proof that. at least in principle there can be a link between DU inhalation and the insurgence of leukaemia.
- Some other relevant aspect remain to be analysed and discussed. The authors of this paper declare themselves not competent to give an answer. One of these is the cause-effect relationship between the exposure to DU and the death for leukaemia observed for many Italian (and not only Italian) soldiers during the last weeks.

6. "Old" DU (decay products at equilibrium.

We have seen that the efficiency of the separation of nuclides, during the various phases of the process of enrichment of uranium, may not be perfect and may not exactly evaluated.
We therefore have calculated the same EDE considering the composition of DU as shown in tables 1a and 1b ("old" uranium) in which all the non-gaseous radio nuclides originated from the decay of U238, U235 and U234 are considered. The main difference is the presence of Th230, which now is taken into consideration.

The results are shown in table 5 for the total dose.

Table 5 – EDE for 1-hour inhalation of DU, distance 1 km, Sv/g DU released

	"new" uranium	"old" uranium

	Composition as in Tab. 2a/2b	Composition as in Tab. 1a/1b
Clean DU	**6.9e-8**	**1.0e-7**
Dirty DU	**1.1e-7**	**1.7e-7**

We can see as in the case of old DU the dose is increased by about 45% for clean DU and by about 70% for dirty DU.

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