Military use of depleted uranium: assessment of prolonged population exposure

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Abstract

This work is an exposure assessment for a population living in an area contaminated by use of depleted uranium (DU) weapons. RESRAD 5.91 code is used to evaluate the average effective dose delivered from 1, 10, 20 cm depths of contaminated soil, in a residential farmer scenario. Critical pathway and group are identified in soil inhalation or ingestion and children playing with the soil, respectively. From available information on DU released on targeted sites, both critical and average exposure can leave to toxicological hazards; annual dose limit for population can be exceeded on short-term period (years) for soil inhalation. As a consequence, in targeted sites cleaning up must be planned on the basis of measured concentration, when available, while special cautions have to be adopted altogether to reduce unaware exposures, taking into account the amount of the avertable dose.
1 Introduction

Munitions containing depleted uranium (DU) have been used by NATO and US forces during the war operations in Iraq (1991), Bosnia (1994), Kosovo and Serbia (1999). Recently some information on 112 sites targeted by DU weapons in Kosovo has been supplied by NATO to the United Nation Environmental Program Balkans Task Force (UNEP BTF); on November 2000 measurements to detect contamination have been undertaken by a UNEP team in 11 among the 112 sites.

Aim of this paper is outlining some aspects of the exposure of people living in an area contaminated by DU, on the basis of official available information and of simulations, looking for main pathways of average and critical exposure.

Individuation of pathways of high exposure could allow to advice to population; average dose assessment, together with measures of DU concentration in soil, will make delimitation of areas to be cleaned up possible.

2 Military use of depleted uranium

The Gulf war against Iraq in 1991 was the first one known where DU rounds have been used in large quantity (approximately 300 tonnes) [1, 2]. The consequences on the health of the Iraqi population and of the US veterans are still under study. DU exposure at the moment is not considered the most probable cause of the Gulf War Syndrome experienced by hundreds thousand veterans [3]; on the other hand, the effects of the DU left over the Iraqi territory are difficult to show, due to the large number of toxic substances dispersed in the environment during the war and the deterioration of the sanitary situation caused by the embargo to which the country is submitted from 1991 (cited work in [4], app.3).

Reports on potential effects on human health and environment from the use of DU have appeared during the last years: studies on risk assessment for the Jefferson Proving Ground, a US facility for testing DU munitions, have been performed [4]; the risk for population for the Kosovo conflict and for the Gulf war has been also considered [4, 5].

DU can be obtained as by-product in the enrichment process of natural uranium for the production of nuclear fuel and for military applications; as
the ore extracted natural uranium, DU is associated to a reduced chain of radioactive isotopes, formed by $^{238}\text{U}$ and $^{235}\text{U}$ decay products having shorter decay times: $^{234}\text{Th}$ (24 days), $^{234}\text{mPa}$ (1.17 min) and $^{234}\text{Pa}$ (6.7 hours), $^{231}\text{Th}$(25.5 hours). DU can also be obtained by the reprocessing of nuclear power plant spent fuel, and so traces of transuranic elements and $^{236}\text{U}$ can be present. According to official information, DU used by the U.S. Department of Defence contains approximately 0.2% of $^{235}\text{U}$ and traces of $^{234}\text{U}$, $^{236}\text{U}$. Following the indications in [7, 8], we will assume the uranium isotopic composition of DU given in Table 1. DU specific activity is in part due to uranium isotopes (14.9 Bq/mg, 36%), and for the residual part to beta emitting short-life decay products (64%); among the transuranic elements official information is available only for $^{239}\text{Pu}$ (2.4 $10^4$ years), whose content is estimated in 11 ppb [9]. DU specific activity is not substantially affected by the declared amount of traces elements.

Metallic uranium has a high density (19 g/cm$^3$), is pyrophoric and cheaper than tungsten, and so has been attractive for U.S. Army for the production of armor piercing ammunition since 1960s. Tungsten alloys have been preferred up 1973, when a DU alloy with 0.75% of titanium (U-3/4Ti) was adopted for ammunition made by a thin cylinder in DU alloy encased with lighter material. Systems of DU weapons are owned or under development in different countries (Saudi Arabia, France, United Kingdom, Israel, Pakistan, Russia, Thailand and Turkey) [8].

Use of DU ammunition causes exposure of people soon and after, because DU is dispersed as aerosol when the projectile strikes a hard target and

Table 1: Assumed depleted uranium composition. $A_i$ is the specific isotopic activity, $A_{DU}$ is the activity concentration per mg of DU.

|      | %  | $T_{1/2}$ (y) | $A_i$ (Bq/mg) | $A_{DU}$ (Bq/mg) |
|------|----|--------------|---------------|-----------------|
| $^{238}\text{U}$ | 99.796 | $4.5 \times 10^9$ | 12.4 | 12.375 |
| $^{235}\text{U}$ | 0.2 | $0.7 \times 10^4$ | 80 | 0.160 |
| $^{234}\text{U}$ | 0.001 | $2.5 \times 10^5$ | $2.3 \times 10^5$ | 2.300 |
| $^{236}\text{U}$ | 0.003 | $2.3 \times 10^7$ | $2.4 \times 10^4$ | 0.072 |
| $\sum U$ | 100 | | | 14.907 |
then falls out on a limited area. Contamination of all environmental matrices takes place and health effects on people living nearby must be taken in account, both for toxicological damage and for radiological risk. Among different isotopes present in DU as declared, $^{238}U$, $^{234}U$ and $^{235}U$ are of concern in risk assessment. For chemical hazard, kidney is identified as the target organ, whatever the path of assumption. Due to prevalent short-range emitted radiation, the risk associated with exposure to ionizing radiation mainly derives from ingestion and inhalation of radioactive material; external irradiation from soil is less relevant.

3 Dispersion of DU in the environment and exposure of the population

DU contained in projectiles, spread out as aerosol in air after striking the target, falls out producing environmental and food chain contamination. Possible occurring of chemical hazard and entity of radiation dose must be assessed for people living in the area, taking into account both average and critical group exposure.

DU concentration in the soil is the starting point; while waiting for measurements of contamination in Iraq, Bosnia, Kosovo and Serbia, we present computed radiation doses and associated concentrations for different contaminated soil thickness, as soil mixing will extend the initial superficial deposition to underlying layers in not undisturbed areas. Available soil measured DU concentrations in contaminated sites that we are aware of, are the following:

- at Jefferson Proving Ground area an average $\sum U$ concentration of 318 Bq/kg was reported; more recently a lower and an upper bound of the concentration ranging from 592 Bq/kg to 13690 Bq/kg was also measured;
- among the areas where the US personnel lived in the Gulf region (outside Iraq) the highest DU concentration (433 Bq/kg) was measured in the Iraqi Tank Yard (the area where captured Iraqi equipment is stored in Kuwait);
in some sample analyzed by the RFY scientists a specific activity of $^{238}\text{U}$ up to $2.35 \times 10^5 \text{ Bq/kg}$ was detected \cite{13}.

Following the hypothesis assumed in the BTF report, we have assumed as a reference value a DU contamination of $1000 \text{ Bq/kg}$ of soil over an area of $A = 10000 \text{ m}^2$, in the hypothesis of $10 \text{ kg}$ of DU entirely dispersed in the impact as aerosol of uranium oxides, contaminating $1 \text{ cm}$ of soil. With the composition given in Table 1 initial activities per $\text{kg}$ of soil for $^{238}\text{U}$, $^{235}\text{U}$, $^{234}\text{U}$ and $^{236}\text{U}$ are respectively $830 \text{ Bq}$, $11 \text{ Bq}$, $154 \text{ Bq}$ and $5 \text{ Bq}$.

Average effective dose is conservatively assessed using the residential farmer scenario. The following pathways are considered: external irradiation from soil, inhalation from resuspended dust, ingestion of contaminated soil and water, ingestion of plants and animal products grown in site and ingestion of fish grown in a pond contaminated by groundwater. Different pathways are considered for plant contamination due to first root uptake (water independent) and due to secondary root uptake from use of contaminated water (water dependent). Radon inhalation is excluded. RESRAD 5.91 \cite{14} code is used, all parameters default except for the ones given in Table 2. Estimates of dose to individuals and population for risk in contaminated sites have been performed by EPA employing primarily the code RESRAD (for related work see \cite{15, 16}).

RESRAD default libraries values have been corrected to give effective dose \cite{17} rather than equivalent effective dose \cite{18}: due to the algorithm used by RESRAD, anyway, values for external irradiation $E_G$ in Tables 4 and 5 have been impossible to modify, and are approximate by $10\%$ maximum defect.

In Tables 3, 4, 5 and 6 we show average annual effective doses and corresponding DU concentrations in water and vegetables for three different soil thickness, respectively $1 \text{ cm}$, $10 \text{ cm}$ and $20 \text{ cm}$. The following quantities are given at different times, from the first year to about two hundred years after maximum dose, for main pathways: the total dose ($E_{\text{tot}}$), the dose from external irradiation from the soil ($E_G$), from inhalation of contaminated dust ($E_I$), from consumption of edible plants (water independent $E_P$, water dependent $E_{wP}$) and of water ($E_{H_{2}O}$).

The dependence of $t_{\text{max}}$ and $E_{\text{max}}$ on some hydrogeological parameters, mainly affecting the water dependent pathways, is shown in Table 7 and 8. The maximum value of the dose is not much affected by most of the parameters considered in Table 8 except $K_d$. This parameter is defined as
Table 2: RESRAD parameters different from the default value.

|                         | this paper | RESRAD def |
|-------------------------|------------|------------|
| indoor time fraction    | 0.6        | 0.5        |
| outdoor time fraction   | 0.2        | 0.25       |
| exposure duration       | 50 years   | 30 years   |
| well pump intake depth  | 3 m        | 10 m       |
| drinking water intake   | 730 l/y    | 510 l/y    |

the ratio of the mass of solute species observed in the solids per unit of dry mass of the soil to the solute concentration in the liquids. A wide range has been observed for uranium $K_d$ values [19]. For largest value of $K_d$ the DU is retained in surface and does not reach at least within the first 1000 years the watertable. A measurement of the local value of this parameter is therefore necessary to reduce the uncertainty on the dose assessment.

Strong dependence of maximum inhalation dose has been found, as expected, on the dust loading parameter, as shown in Table 3.

As already outlined, presented doses and concentrations have been obtained from an average value of soil contamination, in order to assess the average exposure of population. Whatever the average value considered, anyway, highly inhomogeneous soil concentrations must be expected in the contaminated area, both for sparse aerosol deposition and for oxidation of DU fragments: concentrations up to 12% in weight have been reported [20]. In order to assess the dose to critical population group, this must be taken in account, especially if inhalation of soil was the critical pathway: inhalation of 0.1 g of soil with maximum reported DU contamination, equal to 12 mg DU, corresponds to 1.44 mSv; ingestion of 1 g of soil, equal to 120 mg DU, corresponds to 0.08 mSv.

A scenario, in which permanence in dusting air and ingestion of soil are possible, is the one for children playing with soil. From the presented dose assessment and considerations children playing with soil may be identified as the critical population group, with inhalation and/or ingestion of contaminated soil as critical pathway. Evidently, average and critical doses are somehow competitive, because the higher fraction of DU is dispersed as aerosol, the lower part of it can rest in soil as fragment, being presence of fragments
Table 3: Effective doses (µSv) for contaminated soil thickness 1 cm. \( E_{\text{tot}} \), \( E_G \), \( E_I \), \( E_P \), \( E_{H_2O} \), \( E_P^w \) are the total dose, the ground, inhalation, plant (water independent), water, plant (water independent) doses. The initial contamination is assumed of 1000 Bq/kg over an area of \( A = 10000 \) m². The symbol - means doses less than 1 µSv. All not specified parameters as in Table 2.

| \( t(y) \) | \( E_{max} \) | \( E_G \) | \( E_I \) | \( E_P \) | \( E_{H_2O} \) | \( E_P^w \) |
|---|---|---|---|---|---|---|
| 0  | 4  | 4  | -  | -  | -  | -  |
| 1  | 3  | 3  | -  | -  | -  | -  |
| 3  | -  | -  | -  | -  | -  | -  |
| 300| -  | -  | -  | -  | -  | -  |
| 485| 4  | -  | -  | -  | 4  | -  |
| 500| 4  | -  | -  | -  | 4  | -  |
| 700| -  | -  | -  | -  | -  | -  |

the main cause of hot spots in soil contamination.

It must be outlined that the amount of DU considered in the simulation corresponds to 37 A-10 /GAU-8 ammunitions. According to the available information, a much larger number of projectiles has been fired on each site (between 50 and 2320, average 300) and up to now unknown is the extension of targeted sites. Both for average and critical exposure, anyway, more realistic dose assessment will be possible only when measured contamination data will be known, scaling the values in the tables for the appropriate factor.

Increment of inhalation dose attributable to \(^{239}\text{Pu}\) presence in DU is officially estimated in 14% [9]: with 11 ppb of \(^{239}\text{Pu}\) in DU RESRAD gives a maximum dose increment of 0.6%.

4 Normative and recommendations framework

Before discussing compliance of average assessed doses and exposure with international standards set to prevent from toxicological damage and limit ionizing radiation risk, we shortly line out an aspect relative to radioprotection system, maybe useful even in wider considerations on risk.
Due to accepted linear-no-threshold model for effects produced by ionizing radiation, justification of a practise has to be the first one posed, that is if the population exposure from military use of DU is justified or not. Comparison between dose estimates in such a scenario and dose limits and dose constraints stated by regulations is anyway useful, for a quantitative perception of risk. In order to assess the need for remediation in contaminated areas, once again the question of justification has to be considered; specific reference levels, linked to the avertable annual dose, have to be defined by national authorities. ”Generic reference levels ... should be used with great caution” and their use ”should not prevent protective actions from being taken to reduce ... dominant components [of existing annual dose]” [21]. We next report a comment to assessed doses, comparing them with radiological and toxicological reference values, in order not to hold the question narrowed to exceeding of dose limits.

Values of annual dose in Tables 3, 4 and 5 show the same temporal shape, with an initial prevalent dose from irradiation by soil and a maximum from ingestion of contaminated drinking water occurring after about five hundreds years, when contamination reaches the aquifer serving the population. Maximum dose, progressively increasing as inventory of DU increases, is always lower than annual population limit (1 mSv/y), starts to be comparable with

| $t(y)$ | $E_{max}$ | $E_{CG}$ | $E_I$ | $E_P$ | $E_{H_2O}$ | $E_{P}^{w}$ |
|-------|-----------|----------|------|------|------------|------------|
| 0     | 18        | 15       | -    | 1    | -          | -          |
| 1     | 17        | 14       | -    | 1    | -          | -          |
| 3     | 15        | 12       | -    | 1    | -          | -          |
| 10    | 9         | 8        | -    | 1    | -          | -          |
| 30    | 2         | 2        | -    | -    | -          | -          |
| 100   | -         | -        | -    | -    | -          | -          |
| 300   | -         | -        | -    | -    | -          | -          |
| 486   | 44        | -        | -    | 41   | 2          |            |
| 500   | 44        | -        | -    | 41   | 2          |            |
| 700   | -         | -        | -    | -    | -          | -          |
Table 5: Effective doses (µSv) for contaminated soil thickness 20 cm. All not specified parameters as in Table 3.

| $t(y)$ | $E_{max}$ | $E_G$ | $E_I$ | $E_{P1}$ | $E_{H2O}$ | $E_{P2}$ |
|-------|-----------|-------|-------|----------|------------|----------|
| 0     | 24        | 19    | 1     | 2        | -          | -        |
| 1     | 23        | 18    | 1     | 2        | -          | -        |
| 3     | 21        | 17    | 1     | 2        | -          | -        |
| 10    | 17        | 13    | -     | 2        | -          | -        |
| 30    | 8         | 7     | -     | 1        | -          | -        |
| 100   | 1         | 1     | -     | -        | -          | -        |
| 300   | -         | -     | -     | -        | -          | -        |
| 499   | 87        | -     | -     | -        | 82         | 4        |
| 700   | 1         | -     | -     | -        | 1          | -        |

Table 6: DU concentrations in the water $C_{H2O}$ and in the edible plants (water dependent) $C_{P}^{w}$ at the maximum dose time.

|         | $C_{H2O}$ (Bq/l) | $C_{P}^{w}$ (Bq/kg) |
|---------|------------------|---------------------|
| 1 cm    | 1.11             | 1.48                |
| 10 cm   | 1.15             | 1.85                |
| 20 cm   | 2.25             | 3.74                |

EPA cleanup limit criterion (150 µSv/y, [22]) for 20 cm depth. Exceeding of dose constraint of 0.1 mSv/y indicated in [21] for long-lived isotopes may not be excluded. This in general happen only after long times, due to the low mobility of the uranium oxides (the mean transit times for insoluble uranium in the top 10 cm of soil range from 7.4 to 15.4 years with an average of 13.4 years [23]; soluble forms have a mean transit times of one month). At the maximum dose time concentration of DU in the water reaches the provisional value of WHO guideline for drinkable water (0.05 Bq/l [24]) already for 1 cm depth. The concentration of DU in leafy vegetables at time of maximum dose ranges from 2 to 4 Bq/kg; no derived limit is defined for consumption of dietary parts.
Table 7: Effective doses for contaminated soil thickness 10 cm, unsaturated zone thickness 3.90 m, for different values of the well pump intake depth (WPID). ($C_{H2O}$ and $C_w^P$ are the concentrations of DU in the water and in the plants (water dep)). All not specified parameters as in Table 2.

| WPID(m) | $t_{max}$(y) | $E_{max}$(µSv) | $C_{H2O}$(Bq/l) | $C_w^P$(Bq/kg) |
|---------|--------------|----------------|-----------------|----------------|
| 1       | 398          | 103            | 2.7             | 4.5            |
| 2       | 417          | 65             | 1.7             | 2.8            |
| 4       | 564          | 33             | 0.7             | 1.4            |

Table 8: Contaminated soil thickness 10 cm. All not specified parameters as in Table 2.

|                  | $t_{max}$(y) | $E_{max}$(µSv) |
|------------------|--------------|----------------|
| prec.rate $(0.9 - 1.1)m$ | 537 - 435   | 43.0 - 44.1    |
| watershed area $(10^6 \pm 10^2)m^2$ | 486         | 43.6           |
| well pumping rate $(200 - 300)m^3/y$ | 486         | 43.6           |
| distrib.coeff. $K_d$ $(20 - 100)cm^3/g$ | 215 - 0     | 118 - 19       |

Inhalation of highly contaminated soil may lead to exceeding of annual dose limit, with possible occurring of toxicological damage: maximum allowed concentration in air for workplaces stated by NRC, 45 µg/m³ for soluble and 200 µg/m³ for insoluble uranium forms, would be exceeded if dust loading was more than 1700 µg/m³, a high but not extreme value. Less important seems ingestion of contaminated soil, due to the lower value of dose conversion factor with respect to the inhalation one. Anyway, ingestion of 1 g maximum contaminated soil would result in 120 mg DU ingestion, when maximum daily ingestion of uranium, due to toxicological effects, was stated in 150 mg by Italian legislation till year 2000.
Table 9: Average dose from inhalation at $t = 0$ for different values of the dust loading parameter. Contaminated soil thickness 10 cm. All not specified parameters as in Table 2.

| inhal. dose (µSv) | 100 µg/m$^3$ | 1 mg/m$^3$ | 5 mg/m$^3$ |
|-------------------|--------------|------------|------------|
|                   |             | 3          | 16         |

5 Conclusions

DU contained in projectiles, spread out in air after striking the target, falls out producing environmental and food chain contamination. Possible occurring of chemical hazard and entity of radiation dose must be assessed for different kind of exposure of people living in the area, taking into account both average and critical group exposure. While waiting for measurements of contamination in Iraq, Bosnia, Kosovo and Serbia, we have computed radiation doses and concentrations for different contaminated soil thickness, as soil mixing will extend the initial superficial deposition to underlying layers in not undisturbed areas.

In order to assess the average exposure of population, doses and concentrations have been obtained from an average value of soil contamination. For the individuation of the critical group inhomogeneous soil concentration has been considered. The presented dose assessment suggests a short term exposure due to inhalation and/or ingestion of contaminated soil and a long term exposure due to ingestion of contaminated water and food; the propagation of the superficial contamination to the watertable critically depends on various hydrogeological parameters to be evaluated on the site.

In sites targeted by DU munitions special cautions have to be adopted to reduce unaware exposures and cleanup must be planned on the basis of the measured concentrations.

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