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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 the use of depleted uranium (DU) weapons. RESRAD 5.91 code was used to evaluate the average effective dose at depths of 1, 10, 20 cm of contaminated soil, in a residential farming scenario. Critical pathways and groups are identified in soil inhalation and ingestion; critical group is identified in children playing with the soil. From the available information on DU released at targeted sites, both critical and average exposure can produce toxicological hazards. The annual dose limit for the population can be exceeded within a few years from DU deposition for soil inhalation. As a result, clean up at targeted sites must be planned on the basis of measured concentration, when available, while special measures must be adopted anyway to reduce unaware exposures.

Keywords: Depleted uranium; Exposure assessment

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

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was supplied by NATO to the United Nations Environmental Program Balkans Task Force (UNEP BTF). In November 2000 measurements to detect contamination were undertaken by a UNEP team at 11 of the 112 sites.

The aim of this paper is to outline some aspects of the exposure of people living in an area contaminated by DU, based on officially available information and simulations and to determine main pathways for average and critical exposure.

Isolation of high exposure pathways would allow specific advice to be given to the population. Average dose assessment, together with measures of DU concentration in the soil, would 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 known conflict where DU rounds were used in large quantity (approximately 300 DU tons) (Table F in US DoD, 1998). The health consequences on the Iraqi population and 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 of thousands of veterans (Hodgson and Kipen, 1999); on the other hand, the effects of the DU left on Iraqi territory are difficult to prove, due to the large number of toxic substances dispersed in the environment during the war and the deterioration of sanitation caused by the *embargo* inflicted on the country since 1991 (cited work in app. 3 of UNEP BTF, 1999).

Reports on the potential effects on human health and the environment from the use of DU have appeared during recent years. These include studies on the risk assessment for the Jefferson Proving Ground, a US facility for testing DU munitions (Ebinger, 1998) and the risk assessment for the population, as a result of the Kosovo conflict and the Gulf war (UNEP BTF, 1999; Fetter & von Hippel, 1999).

DU can be obtained as a by-product in the enrichment process of natural uranium in the production of nuclear fuel and for military applications. As the ore extracted natural uranium, DU is associated with a reduced chain of radioactive isotopes, formed by ^{238}U and ^{235}U β-emitter decay products having shorter decay times: ^{234}Th (24 days), ^{234}Pa (1.17 min) and ^{234}Pa (6.7 h), ^{231}Th (25.5 h). DU can also be obtained as a by-product in the reprocessing of nuclear power plant spent fuel, and therefore traces of transuranic elements and ^{236}U can be present. According to official information, DU used by the US Department of Defence contains approximately 0.2% ^{235}U and traces of ^{234}U, ^{236}U. Following the indications of US AEPI (1995) and Harley et al. (1999), we will assume the uranium isotopic composition of DU given in Table 1.

DU specific activity is due in part to alpha emitting uranium isotopes (14.9 Bq /mg, 36%); the remaining part is due to beta emitting short-life decay products, mentioned above, (64%). Among the transuranic elements, official information is available only for ^{239}Pu ($2.4\ 10^4$ years), whose content is estimated at 11 ppb (US DoE, 2000). DU specific activity is not substantially affected by the declared amount of trace elements.

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}U	99.796	$4.5\ 10^9$	12.4	12.375
^{235}U	0.2	$0.7\ 10^9$	80	0.160
^{234}U	0.001	$2.5\ 10^5$	$2.3\ 10^5$	2.300
^{236}U	0.003	$2.3\ 10^7$	$2.4\ 10^3$	0.072
Σ_U	100			14.907

Metallic uranium has a high density (19 g/cm^3), is pyrophoric and cheaper than tungsten, and so has been attractive to the US Army for the production of armor piercing ammunition since the 1960s. Tungsten alloys were preferred up to 1973, until 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. DU weapons systems are owned or under development in different countries (Saudi Arabia, France, United Kingdom, Israel, Pakistan, Russia, Thailand and Turkey) (Harley et al., 1999).

The use of DU ammunition causes exposure to people both immediately and afterwards, because DU is dispersed as an aerosol when the projectile strikes a hard target, and then falls out on a limited area (US AEPI, 1995). Contamination of all environmental matrices takes place and health effects on people living nearby must be taken into account, both for toxicological damage and for radiological risk. Among the different isotopes present in DU as declared, ^{238}U, ^{234}U and ^{235}U are of concern in risk assessments. As far as chemical hazard is concerned, the kidney has been identified as the target organ, whatever the assumed pathway of assumption (BEIR V, 1990). Due to the prevalent short-range radiation emitted, the risk associated with exposure to ionizing radiation is mainly derived 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 spreads out as an aerosol in the air after striking the target; subsequent fall out produces environmental and food chain contamination. The possibility of chemical hazard and the amount of radiation dose must be assessed for the people living in the area, taking into account both the average and critical group exposure.

DU concentration in the soil is the starting point. While waiting for systematic 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 disturbed areas. Available measures of DU concentrations in the soil of contaminated sites that we are aware of, are the following:

- At the Jefferson Proving Ground area, an average ΣU concentration of 318 Bq/kg was reported (Ebinger and Hansen, 1994); more recently a lower and an upper bound of the concentration ranging from 592 Bq/kg to 13,690 Bq/kg was also measured (Ebinger, 1998);
- Among the areas where US personnel lived in the Gulf region (outside Iraq) the highest DU concentration (433 Bq/kg) was measured in the Iraqi Tank Yard (the areas in Kuwait where captured Iraqi equipment is stored) (US DoD, 2000);
- In some samples analyzed by scientists of Federal Republic of Yugoslavia, a specific activity of ^{238}U up to 2.35 10^5 Bq/kg was detected (Petković et al., 2000).

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

The average effective dose is assessed conservatively using the residential farming 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 on the 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 the use of contaminated water (water dependent). Radon inhalation is excluded. RESRAD 5.91 (ANL, 1989) code is used, all parameters default except for the ones given in Table 2. Estimates of dose to individuals and population in contaminated sites have been performed by EPA employing primarily the code RESRAD (for related works see Wolbarst et al., 1996 and Wood et al. (1999).

RESRAD default library values have been corrected to give the effective dose (ICRP, 1990), rather than the equivalent effective dose (ICRP, 1977). Due to the algorithm used by RESRAD, however, values for external irradiation E_G in Tables 4 and 5 were impossible to modify, and have approximately 10% maximum defect.

Tables 3, 4, 5 and 6 show average annual effective doses and corresponding DU concentrations in water and vegetables for three different soil thickness, 1 cm, 10 cm and 20 cm respectively. The following quantities are given at different times,

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

Table 4
Effective doses (μSv) for contaminated soil thickness 10 cm. E_{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 to be 1000 Bq/kg over an area of A=10000 m^2. The symbol (–) means doses less than 1 μSv. All RESRAD parameters default except those in Table 2

$t(y)$	E_{tot}	E_G	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. E_{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 to be 1000 Bq/kg over an area of A=10000 m^2. The symbol (–) means doses less than 1 μSv. All RESRAD parameters default except those in Table 2

$t(y)$	E_{tot}	E_G	E_I	E_P	E_{H_2O}	E_P^w
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	–

from the first year to about 200 years after the time of maximum dose, for main pathways: the total dose (E_{tot}), the dose from external irradiation from the ground (E_G), from inhalation of contaminated dust (E_I), from consumption of edible plants (water independent E_P, water dependent E_P^w) and of water (E_{H_2O}).

The dependence of t_{max} and of the total dose at t_{max} (E_{max}) on some hydrogeological parameters which affects mainly the water dependent pathways, is shown in Tables 7 and 8. The maximum value of the dose is not greatly affected by most of the parameters considered in Table 8 except K_d. This parameter is defined as 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 (Sheppard and Thibault, 1990). For the largest value of K_d the DU

Table 3
Effective doses (μSv) for contaminated soil thickness 1 cm. E_{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 to be 1000 Bq/kg over an area of A=10000 m^2. The symbol (–) means doses less than 1 μSv. All RESRAD parameters default except those in Table 2

$t(y)$	E_{tot}	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	–	–	–	–	–	–

Table 6
DU concentrations in the water C_{H_2O} and in the edible plants (water dependent) C_P^w at the maximum dose time

	C_{H_2O}(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

Table 7
Maximum effective dose, E_{max}, for contaminated soil thickness 10 cm, unsaturated zone thickness 3.90 m, for different values of the well pump intake depth (WPID). (C_{H_2O} and C_P^w are the concentrations of DU in the water and in the plants (water dep)). All RESRAD parameters default except those in Table 2

WPID (m)	t_{max} (y)	E_{max} (μSv)	C_{H_2O}(Bq/l)	C_P^w (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
Maximum effective dose, E_{max}, varying some hydrogeological parameters for contaminated soil thickness 10 cm. All RESRAD parameters default except those in Table 2

	t_{max} (y)	E_{max} (μSv)
Prec. rate (0.9–1.1) m	537–435	43.0–44.1
Watershed area ($10^6 \pm 10^5$) m^2	486	43.6
Well pumping rate (200–300) m^3/y	486	43.6
Distributed coefficient K_d (20–100) cm^3/g	215– 0	118–19

is retained in surface and does not reach the watertable, at least within the first 1000 years. A measurement of the local value of this parameter is therefore necessary to reduce the uncertainty concerning dose assessment.

As expected, a strong dependence of the maximum inhalation dose on the dust loading parameter was found, as shown in Table 9.

As already outlined, in order to assess the average exposure of population, presented doses and concentrations were obtained from an average value of soil contamination. Whatever average value is considered, however, 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 previously been reported (Ebinger et al., 1990). In order to assess the dose to a critical population group, this must be taken into account, especially if inhalation of soil was the critical pathway: inhalation of 0.1 g of soil with the maximum reported DU contamination, equals 12 mg DU and corresponds to 1.44 mSv; ingestion of 1 g of soil, equal to 120 mg DU, corresponds to 0.08 mSv.

A scenario which is possible, the permanence in dusting air and the ingestion of soil, is the one of 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 the critical pathway. Evidently, average and critical doses are somewhat competitive, because the higher fraction of DU is dispersed as an aerosol, the lower fraction rests in the soil as fragments (the main cause of hot spots in soil contamination).

It must be underscored 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 was tired on each site (between 50 and 2320, average 300) and up to now the extent of targeted sites is unknown. For both average and critical exposure, however, more realistic dose assessment will only be possible when measured contamination data is known, and the values in the tables can be scaled for the appropriate factor.

The increment of the inhalation dose attributable to the presence of ^{239}Pu in DU is officially estimated to be 14% (US DoE, 2000): with 11 ppb of ^{239}Pu in DU, RESRAD gives a maximum dose increment of 0.6%.

4. Normative and recommendations framework

Before discussing the compliance of average assessed doses and exposure with international standards set to prevent toxicological damage and limit ionizing radi-

Table 9
Average dose from inhalation at t=0 for different values of the dust loading parameter. Contaminated soil thickness 10 cm. All RESRAD parameters default except those in Table 2

	100 μg/m^3	1 mg/m^3	5 mg/m^3
Inhal. dose (μSv)	–	3	16

ation risk, we briefly outline an aspect relative to the radioprotection system, which could be useful within even wider risk considerations.

The first question posed from the accepted linear-no-threshold model for effects produced by ionizing radiation, has to be justification of practice, that is, is the population exposure from military use of DU justified or not. Comparison between dose estimates in such a scenario and dose limits and dose constraints stated by regulations is useful anyway, 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]" (ICRP, 2000).

In order to identify such protective actions, we believe that assessed doses should be compared with different radiological and toxicological reference values, without reducing the question to the exceeding of radiological 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 hundred years, when, contamination reaches the aquifer serving the population. Maximum dose, which progressively increases as the inventory of DU increases, is always lower than the annual population limit (1 mSv/y), and starts to be comparable with EPA cleanup limit criterion (150 μSv/y, (US EPA, 1997)) for 20 cm depth. Exceeding the dose constraint of 0.1 mSv/y indicated in ICRP (2000) for longlived isotopes may not be excluded. This generally happens only after a long time, due to the low mobility of the uranium oxides (the mean transit times for insoluble uranium in the top 10 cm of soil ranges from 7.4 to 15.4 years, with an average of 13.4 years (Killough et al., 1999); soluble forms have a mean transit time of one month). At the maximum dose, concentration of DU in the water reaches the provisional value of WHO guidelines for drinkable water (0.05 Bq/l (WHO, 1998)) already at 1 cm depth of contamination. The concentration of DU in leafy vegetables at the time of maximum dose, ranges from 2 to 4 Bq/kg*;* no derived limit is defined for consumption of dietary parts.

Inhalation of highly contaminated soil may result in exceeding the annual dose limit, with the possible occurrence of toxicological damage: the maximum concentration in air for workplaces as stated by the NRC (45 $\mu g/m^3$ for soluble and 200 $\mu g/m^3$ for insoluble uranium forms), would be exceeded if dust loading was more than 1700 $\mu g/m^3$, a high but not extreme value. Less important seems the ingestion of contaminated soil, due to the lower dose conversion factor with respect to inhalation. Nonetheless, ingestion of 1 g maximum contaminated soil would result in ingestion of 120 mg DU, with the maximum daily ingestion of uranium from toxicological effects stated to be 150 mg by the Italian legislature until the year 2000.

5. Conclusions

DU contained in projectiles is spread out in the air after striking the target and subsequently falls out, producing environmental and food chain contamination. Possible occurrence of a chemical hazard and the amount of the radiation dose must be assessed for various kinds of exposure to people living in the area, taking into account both average and critical group exposure. While waiting for systematic measurements of contamination in Iraq, Bosnia, Kosovo and Serbia, we computed radiation doses and concentrations for different contaminated soil thicknesses, as soil mixing will extend the initial superficial deposition to underlying layers in disturbed areas.

In order to assess the average exposure of the population, doses and concentrations were obtained from an average of soil contamination values. For the isolation of the critical group a inhomogeneous soil concentration was considered.

The presented dose assessment suggests a short term exposure due to inhalation and/or ingestion of contaminated soil (the critical group was identified as children playing with the soil) and 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 which need to be evaluated at the site.

In sites targeted by DU munitions, special measures have to be adopted to reduce unaware exposure and cleanup must be planned based on the measured concentrations.

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