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Depleted uranium residual radiological risk assessment for Kosovo sites

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Abstract

During the recent conflict in Yugoslavia, depleted uranium rounds were employed and were left in the battlefield. Health concern is related to the risk arising from contamination of areas in Kosovo with depleted uranium penetrators and dust. Although chemical toxicity is the most significant health risk related to uranium, radiation exposure has been allegedly related to cancers among veterans of the Balkan conflict. Uranium munitions are considered to be a source of radiological contamination of the environment. Based on measurements and estimates from the recent Balkan Task Force UNEP mission in Kosovo, we have estimated effective doses to resident populations using a well-established food-web mathematical model (RESRAD code). The UNEP mission did not find any evidence of widespread contamination in Kosovo. Rather than the actual measurements, we elected to use a desk assessment scenario (*Reference Case*) proposed by the UNEP group as the source term for computer simulations. Specific applications to two Kosovo sites (Planeja village and Vranovac hill) are described. Results of the simulations suggest that radiation doses from water-independent pathways are negligible (annual doses below 30 μSv). A small radiological risk is expected from contamination of the groundwater in conditions of effective leaching and low distribution coefficient of uranium metal. Under the assumptions of the *Reference Case*, significant radiological doses (>1 mSv/year) might be achieved after many years from the conflict through water-dependent pathways. Even in this worst-case scenario, DU radiological risk would be far overshadowed by its chemical toxicity.

Keywords: Depleted uranium; Radioactive contamination; RESRAD code; Radiological risk

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1. Introduction

The recent conflict in Yugoslavia is a source of public concern due to the radiological and toxic hazards posed by depleted uranium (DU) weapons used by NATO forces. DU is used in armor-piercing incendiary (API) anti-tank munitions, such as those aboard A-10 Warthog jets. About 31,000 API rounds were allegedly fired in Kosovo.

Concern about the health risks related to DU weapons has been raised, based on the well-known chemical and radiological toxicity of uranium. The main health threat related to uranium use is chemical toxicity (Keith et al., 1999). Like many other heavy metals, uranium is toxic to humans and animals, with the kidney being the target organ (Leggett, 1989).

Despite its prominent chemical toxicity, and although review of scientific literature addressing occupational exposure to uranium provides little evidence of cancer risk, most of the concern for the health of soldiers and civilians has been related to DU radioactivity (Harley et al., 1999; Fulco et al., 2000; McDiarmid, 2001; Priest, 2001). Committed effective dose per unit of intake of DU is 0.12 mSv/mg for inhalation (adults, type-S absorption) and 0.67 μSv/mg for ingestion (ICRP, 1996; UNEP, 1999). The dose rate for external exposure is mostly due to β radiation and it is around 2 mSv/h for the skin in contact with a DU penetrator (Fetter and von Hippel, 1999).

Several experimental studies are under way in the Balkans to evaluate the health risk related to DU, including epidemiological studies on cancer deaths and measurements of radioactivity in the soil and water. Careful measurements have been performed by the UNEP (2000) Balkan Task Force Mission. Although the task force did not find any evidence of widespread DU contamination, they point out the possible hazard to groundwater in future years. DU contamination is especially important for resident populations, who are subjected to inhalation of dusts, external exposure from penetrators on the ground, food and water contamination, and so on. All of these pathways are simulated in the RESRAD software (Yu et al., 2001). In a recent paper (Durante and Pugliese, 2002), we used RESRAD to simulate a number of worst-case scenarios with different DU concentrations in soil and water, including extreme conditions with very high DU concentrations in the soil (up to 40 Bq/g) and in the groundwater (up to 50 Bq/l). The purpose of this paper is to provide more realistic theoretical hazard predictions from RESRAD, using UNEP (2000) data and assumptions as the source terms.

Local radioactive hotspots, such as contact with single API rounds, abandoned tanks or targets struck by DU munitions etc., are not considered in these simulations.

2. Materials and methods

2.1. UNEP data

As part of the post-conflict environmental study carried out by the United Nations Environmental Programme (UNEP) in the Balkans, a Depleted Uranium Desk

Assessment Group prepared a preliminary study in October 1999 (UNEP, 1999), including theoretical analyses. During a subsequent field mission to Kosovo from 5 to 19 November 2000, soil, water, and biological samples were collected at 11 sites where DU reportedly had been used during the conflict (UNEP, 2000). Five independent laboratories analyzed the samples. Only low-level radioactive contamination was measured in the samples from Kosovo. However, a possible theoretical scenario was postulated in the UNEP preliminary assessment (UNEP, 1999), and this desk assessment scenario, called *Reference Case*, was re-considered following the field mission (UNEP, 2000). In the *Reference Case*, it is assumed that an attack includes three aircraft and 10 kg of DU. The area affected by subsequent DU contamination, including dust caused by explosions and fire, is assumed to be 1000 m^2, and the activity is distributed 10 cm into the soil. After some time, the area may be cultivated. Possible contamination pathways include inhalation of re-suspended dust, external exposure from solid DU pieces, contamination of the groundwater, grazing animals, and so on.

2.2. RESRAD code

Radiological health risk associated with API contamination of the ground has been estimated by the RESRAD 6.1 computer code (Yu et al., 2001), developed at Argonne National Laboratory. The code is designed for dose and risk assessment based on measured concentrations of radioactive materials in the environment. RESRAD considers 14 exposure pathways: inhalation of dust, radon and volatile chemicals, ingestion of plants, meat, milk, aquatic foods, soil and drinking water, incidental ingestion of water while swimming, external exposure from soil and air immersion, and dermal absorption from swimming, showering and soil contact. RESRAD has been used for over 300 sites in the USA and other countries (e.g. Espegren et al., 1996; Wood et al., 1999; Laniak et al., 1997).

The assumption of uniform distribution of DU in soil is incorrect, because studies of radiological contamination in the soil from impacted DU rounds suggest that dispersion and deposition are localized within 10 m from the hit target (Hanson and Miera, 1977; US Army Corp of Engineers, 1997; UNEP, 2000). However, appropriate codes for predictions based on a non-uniform distribution of DU have not yet been developed, although RESRAD provided reasonable dose estimates for non-uniform DU distribution in the Aberdeen and Yuma API proving grounds (Ebinger et al., 1996). The soil concentration used in this simulation should be the average quantity over the considered area. Although this leads to a general overestimation of the calculated doses, the presence of radioactive hotspots poses a health risk for individuals in direct contact with this highly contaminated area.

Based on the *Reference Case* (UNEP, 1999), we assumed 10 kg DU uniformly distributed in 1000 m^2 and 10 cm depth, i.e. 830 mBq/g soil. Background ^{238}U concentration in the soil ranges between 5 and 125 mBq/g (UNEP, 1999). In all simulations, we assumed that ^{238}U is associated with the 235 and 234 isotopes, according to the isotopic proportions reported in UNEP (1999).

Ingestion parameters, food storage times, etc., were used according to the resident farmer scenario. In this scenario, exposed subjects live in the contaminated area and raise crops and livestock for family consumption. This scenario includes all environmental pathways for on-site or near-site exposure and is expected to result in the highest predicted lifetime dose. To test this hypothesis, we also performed simulations using the industrial worker scenario, where exposed subjects spend 8 h/day in the contaminated zone, but do not ingest meat or milk from livestock raised on the site. As expected, simulation results with this scenario provide consistently lower dose values and are not presented in this paper. Numerical values of the coefficients used in the resident farmer scenario are provided in RESRAD 6 Users' Manual (Yu et al., 2001).

The uncertainty of estimates for water-dependent pathways is large. This is because leakage of DU in the groundwater is strongly affected by soil chemical composition, porosity, humidity, Eh, pH, etc. Uranium that is leached from penetrators and dust particles will be transported in the soil and bedrock as U_2^{2+} ions in the precipitating water. In oxidizing conditions, most of the dissolved uranium ions are in the form of soluble ions and can move from the environment to the living organisms. DU API mostly corrodes into hydrated U(VI) oxides, which exist in solution as the uranyl ion UO_2^{2+}, which is complexed by fluoride at $pH<4$ and by phosphates at $4<pH<7.5$. In basic conditions, dissolved uranium is present predominantly as an uranyl carbonate complex, such as UO_2CO_3. Most of the coefficients relative to the hydrogeological and geochemical characteristics of the contaminated area were derived from two specific Kosovo sites (see below).

2.3. Site data

The *Reference Case* has been used as the source term for a computer simulation of committed radiation dose to the population. Specific hydrogeological and geochemical data from two sites were considered to achieve more realistic simulations, especially for aquatic pathways. In particular, UNEP (2000) describes Planeja village as an area with a few centimeters of soil, groundwater at a depth of 2 m, and limestone rock (hydraulic conductivity from $10^{-5.5}$ to 10^{-9} m/s). The waters are supersaturated with respect to calcite and based on experimental measurements of Carrol and Bruno (1991), a distribution coefficient (K_d) of 1000 is assumed. The minimum distance to village wells is 50 m. The porosity of the rocks is 0.1 and the hydraulic head is 0.5 m.

Vranovic hill, on the other hand, is mostly composed of permeable sands. A minimum value of K_d=4 is estimated by UNEP (2000). The porosity of the rock is 0.3, the hydraulic head is 1 m, and the hydraulic conductivity in silty sand ranges between 10^{-3} and 10^{-7} m/s. The minimum depth of the local well water at the foot of the esker ridge is 2 m.

Simulations with RESRAD show that the aquatic pathways for depleted uranium are mostly dependent on the distribution coefficient K_d (Ebinger, 1998). RESRAD uses a default K_d=50 for uranium. For this reason, we selected these two sites with markedly different K_d values, but used the *Reference Case* as the source term for soil concentration. In fact, little radioactive contamination in soil samples was actually measured by the UNEP (2000) field mission.

3. Results

First we simulated the UNEP (1999) *Reference Case* using the default K_d (50), porosity (0.4), hydraulic conductivity (3×10^{-7} m/s), etc. RESRAD values. As expected, water-dependent pathways were not significant under these conditions. Results for water-independent pathways in the first 50 years following contamination are displayed in Fig. 1. Most of the annual effective dose equivalent was caused by irradiation from the ground, followed by plant, soil, inhalation, milk, and meat pathways. The maximum annual dose from all pathways was 16 μSv, and the integrated committed effective dose in 50 years was 400 μSv.

The simulation of the hydrogeological data from Planeja village using the *Reference Case* assumptions are shown in Fig. 2. Due to the high distribution coefficient in the area, aquatic pathways are negligible in this case. RESRAD indicates that several centuries would be needed to achieve some degree of contamination of the groundwater with these parameters. The integrated 50-year committed effective dose is 820 μSv.

The hydrogeological characteristics of Vranovic hill produced a faster contamination of the groundwater. In particular, RESRAD calculations showed that approximately 15 years would be needed to detect uranium leaching in the water and to achieve significant doses to the resident populations, according to the *Reference Case* hypothesis. Aquatic pathways for the Vranovic hill simulations are shown in Fig. 3. Most of the committed dose is induced by water (drinking, showering, etc.) itself, followed by plants, milk, meat and fish consumption. The maximum annual doses

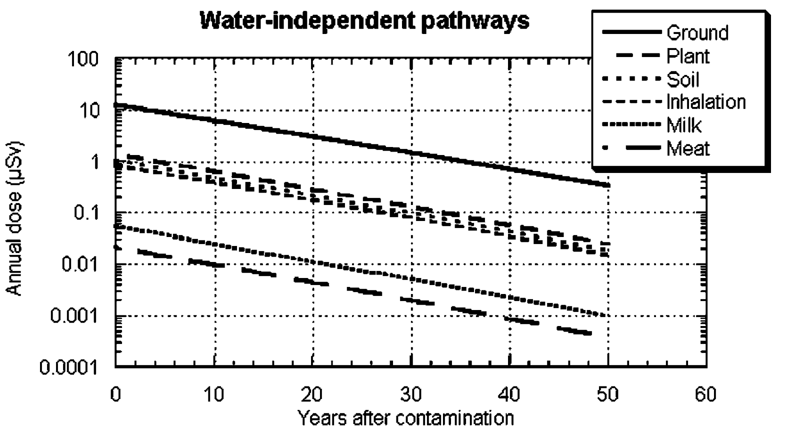

Fig. 1. Estimated annual effective dose equivalent after DU contamination in the UNEP Reference Case. The various contributions of non-aquatic pathways are plotted separately. Water-dependent pathways are negligible in this simulation (hydrogeological data from RESRAD defaults).

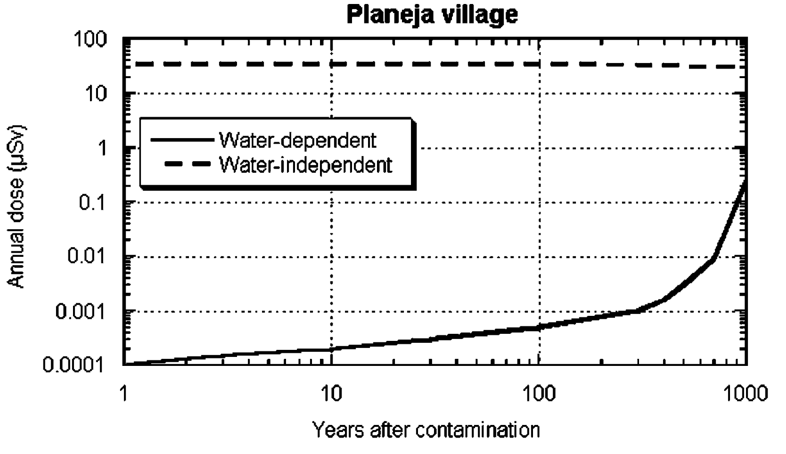

Fig. 2. Estimated annual effective dose equivalent after DU contamination in the UNEP Reference Case, using hydrogeological data from Planeja village in Kosovo. Contributions from water-dependent and -independent pathways are plotted.

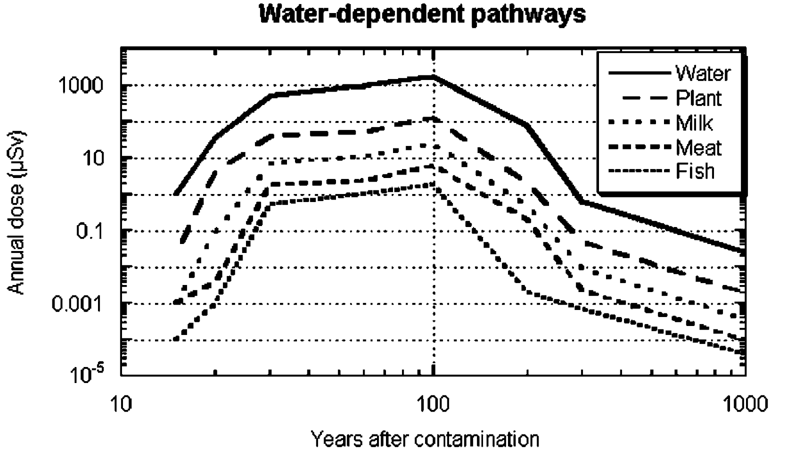

Fig. 3. Annual doses from various water-dependent pathways after DU contamination. Soil concentrations and pathways are based on the UNEP Reference Case, while hydrogeological data are specific for Vranovac hill in Kosovo.

are predicted between 30 and 100 years following soil contamination, and doses will rapidly decrease at later times. A comparison between aquatic and non-aquatic pathways for the Vranovic hill scenario is shown in Fig. 4. In the first few years post-conflict, water-independent pathways largely predominate over aquatic pathways. However, water-dependent pathways become the most important contributor to the annual effective dose after about 20 years, and the effective dose equivalent due to the aquatic pathways can exceed 1 mSv/year between 60 and 100 years after

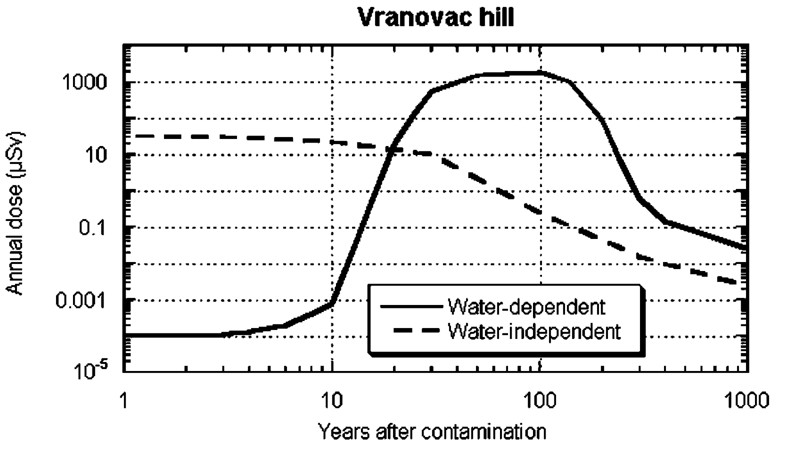

Fig. 4. Estimated annual effective dose equivalent after DU contamination in the UNEP Reference Case, using hydrogeological data from Vranovac hill. Contributions from water-dependent and -independent pathways are plotted.

the contamination. The 50-year committed effective dose for population residents 1 year after the conflict is about 20 mSv.

4. Discussion

UNEP (2000) Balkan Task Force recommendations indicate that the major hazard arising from environmental contamination with DU weapons could be linked to groundwater contamination. Our previous simulations with RESRAD codes (Durante and Pugliese, 2002), using hypothetical worst-case scenarios at different soil and water DU concentrations, demonstrated that water-dependent pathways pose the greatest health hazard to the resident population.

In this paper, we describe a few simulations with more realistic scenarios. Because the UNEP (2000) field mission did not find any evidence of widespread radioactive contamination in Kosovo sites targeted with DU rounds, we have used the UNEP (1999) *Reference Case* as the source term for the RESRAD calculations. In particular, a uniform 830 mBq/g uranium soil concentration in an area of 1000 m^2 was used in all simulations. This should be kept in mind, especially for the results shown in Figs. 2 and 4, where hydrogeological data correspond exactly to the Planeje and Vranoc sites, but contamination data are *Reference Case* assumptions. Therefore, rather than absolute dose values, these simulations provide information about the influence of site characteristics on the expected committed doses due to DU contamination.

All simulations support the hypothesis that water-independent pathways do not give significant contributions to the annual dose to the resident populations. In the simulations performed here, the 50-year committed effective dose never exceeds 1 mSv. This is consistent with our previous RESRAD simulations, where we found that a 50 mSv 50-year committed effective dose can be achieved by water-independent pathways only when DU soil concentrations is greater than 20 Bq/g (Durante and Pugliese, 2002). On the other hand, water-dependent pathways are strongly dependent upon site geochemical and hydrogeological parameters. In particular, a decrease in the distribution coefficient K_d clearly leads to a significant increase in radiological risk from aquatic pathways. Given the Vranovic hill site parameters, it is predicted that any significant risk is likely to occur after about 15–30 years from the war, and might lead to doses which slightly exceed the ICRP (1991) annual dose equivalent recommended limit for the population (1 mSv/year). Nonetheless, even in this scenario, the radiological risk would be insignificant compared to the toxicological risk from uranium ingested through drinking water.

In conclusion, within the limits of the mathematical model used, simulations suggest that no significant radiation doses are expected from water-independent pathways. However, these results support UNEP (2000) recommendations concerning the monitoring of the wells in suspected contaminated areas.

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