

Measurements of daily urinary uranium excretion in German peacekeeping personnel and residents of the Kosovo region to assess potential intakes of depleted uranium (DU)

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Abstract

Following the end of the Kosovo conflict, in June 1999, a study was instigated to evaluate whether there was a cause for concern of health risk from depleted uranium (DU) to German peacekeeping personnel serving in the Balkans. In addition, the investigations were extended to residents of Kosovo and southern Serbia, who lived in areas where DU ammunitions were deployed. In order to assess a possible DU intake, both the urinary uranium excretion of volunteer residents and water samples were collected and analysed using inductively coupled plasma-mass spectrometry (ICP-MS).

More than 1300 urine samples from peacekeeping personnel and unexposed controls of different genders and age were analysed to determine uranium excretion parameters. The urine measurements for 113 unexposed subjects revealed a daily uranium excretion rate with a geometric mean of 13.9 ng/d (geometric standard deviation (GSD)=2.17). The analysis of 1228 urine samples from the peacekeeping personnel resulted in a geometric mean of 12.8 ng/d (GSD=2.60). It follows that both unexposed controls and peacekeeping personnel excreted similar amounts of uranium. Inter-subject variation in uranium excretion was high and no significant age-specific differences were found.

The second part of the study monitored 24 h urine samples provided by selected residents of Kosovo and adjacent regions of Serbia compared to controls from Munich, Germany. Total uranium and isotope ratios were measured in order to determine DU content. $^{235}\text{U}/^{238}\text{U}$ ratios were within $\pm 0.3\%$ of the natural value, and $^{236}\text{U}/^{238}\text{U}$ was less than 2×10^{-7} , indicating no significant DU in any of the urine samples provided, despite total uranium excretion being relatively high in some cases. Measurements of ground and tap water samples from regions where DU munitions were deployed did not show any contamination with DU, except in one sample.

It is concluded that both peacekeeping personnel and residents serving or living in the Balkans, respectively, were not exposed to significant amounts of DU.

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1. Introduction

Uranium is the heaviest naturally occurring element and all its isotopes are radioactive. Its radiotoxicity and chemical toxicity (mainly nephrotoxicity) have been described extensively in the literature (ATSDR, 1999; UNEP, 1999; UNSCEAR, 2000; European Parliament, 2001; WHO, 2001).

Uranium is ubiquitously present in the earth's crust. Depending on the underlying geology, the uranium concentrations in surface soils vary from about 0.1 mg/kg to about 20 mg/kg with a world average of 2.8 mg/kg (UNSCEAR, 2000).

Owing to leaching and re-suspension processes, uranium is also ubiquitous in the aquatic environment and air (Ragnarsdottir and Charlet, 2000). Uranium concentrations in surface and ground water range from 0.1 to over 10,000 $\mu\text{g/l}$ (Santschi and Moneyman, 1989; Orloff et al., 2004) and in air between 0.02 and 0.40 ng/m^3 (average 0.1 ng/m^3) (UNSCEAR, 2000).

It follows from above that all kinds of foodstuff will contain trace amounts of uranium, including tap and mineral water, vegetables, cereals, and table salt (Fisenne et al., 1987; Pietrzak-Flis et al., 2001; Priest, 2001). The mean daily uranium intake was reported to be 1.6 ng from air, 0.11 μg from water and 1.14 μg from food, achieving a total average value of 1.25 $\mu\text{g/d}$ per person (UNSCEAR, 2000). However, intake is highly variable and depends upon local geology, dietary and drinking habits and situational breathing rates (Fisenne et al., 1987; Harley, 1988; Harley et al., 1999; Pietrzak-Flis et al., 2001).

Of the natural uranium ingested with food/drinking water, most is directly excreted in faeces, and the remainder enters the bloodstream following absorption through the wall of the gastrointestinal (GI) tract (Fisher et al., 1983; Larsen et al., 1984; Wrenn et al., 1989; Spencer et al., 1990; Dang et al., 1992b; Harduin et al., 1994; Leggett and Harrison, 1995).

Uranium may also enter the body through the skin or via inhalation. In general, the latter case is more important. In this context, the uranium oxides UO_2 , UO_3 and U_3O_8 are considered to be of main concern (Harley et al., 1999; WHO, 2001). The solubility of these oxides is relatively low so that only about 1% of inhaled uranium is absorbed into the blood stream (ICRP, 1995b; Harley et al., 1999).

After absorption into the bloodstream, either via the lungs or the GI tract, uranium is either rapidly deposited in tissues, mainly in the skeleton and the kidneys, or is excreted in urine (Russell and Kathren, 2004). A detailed description of the uptake, deposition, retention

and clearance of uranium in man can be found elsewhere (ICRP, 1994, 1995a; Stradling et al., 1998; WHO, 2001; Limson Zamora et al., 2002). The total body content of uranium for non-exposed subjects was found to range between 7 μg (Li et al., 2005) and 56 μg (Fisenne et al., 1988).

About two-thirds of the uranium absorbed by the human body is excreted in urine within 24 h of intake (Taylor and Taylor, 1997) and about 90% is eliminated over a period of a few days. Due to its urinary excretion, the concentration of uranium in urine can be used to estimate the size of uranium intakes. Such estimates require both information on the timings of intakes and precise measurements of uranium excretion. Mass spectrometric measurements are the method of choice for the determination of uranium in urine samples, particularly inductively coupled plasma-mass spectrometry (ICP-MS). The use of this method has been recently discussed in detail by Roth et al. (2003). With respect to depleted uranium (DU), particular attention has been given to the improvement of techniques for uranium isotope ratio determination, since $^{235}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ ratios are needed for the assessment of DU in biological samples (Parrish et al., 2006; Oeh et al., in press).

DU is a by-product of the enrichment process used to make nuclear fuel. It contains less ^{235}U (<0.7%, typically 0.2–0.3%) and much less ^{234}U than natural uranium and is about 40% less radioactive. DU has both civilian and military uses (Betti, 2003). The military use is focused on its application in defensive armouring for tanks and as armour-piercing ammunition (Bleise et al., 2003). So far, the use of ammunitions containing DU has been officially confirmed in four military conflicts: Iraq 1991, Bosnia 1994, Kosovo 1999 and Iraq 2003.

During the Kosovo conflict in 1999 about 30 tons of DU munitions were used against military targets (The Royal Society, 2001). Most penetrators, however, are thought to have missed their intended targets. Investigations on the consequences for the environment have already been undertaken (UNEP, 1999; Sansone et al., 2001; UNEP, 2001; Papastefanou, 2002; UNEP, 2002; Jia et al., 2004; Schimmack et al., 2005). On the other hand, when DU penetrators hit hard targets or when DU metal burns, a DU aerosol is produced and this could be inhaled by people and might cause toxic effects if the intake were sufficiently large (Domingo, 2001; Bleise et al., 2003; McDiarmid et al., 2004). The characteristics of such aerosols are described in detail in the published report of the United States Department of Defense CAPSTONE project (US Department of Defense, 2004).

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