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Properties, use and health effects of depleted uranium (DU): a general overview

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Abstract

Depleted uranium (DU), a waste product of uranium enrichment, has several civilian and military applications. It was used as armor-piercing ammunition in international military conflicts and was claimed to contribute to health problems, known as the Gulf War Syndrome and recently as the Balkan Syndrome. This led to renewed efforts to assess the environmental consequences and the health impact of the use of DU. The radiological and chemical properties of DU can be compared to those of natural uranium, which is ubiquitously present in soil at a typical concentration of 3 mg/kg. Natural uranium has the same chemotoxicity, but its radiotoxicity is 60% higher. Due to the low specific radioactivity and the dominance of alpha-radiation no acute risk is attributed to external exposure to DU. The major risk is DU dust, generated when DU ammunition hits hard targets. Depending on aerosol speciation, inhalation may lead to a protracted exposure of the lung and other organs. After deposition on the ground, resuspension can take place if the DU containing particle size is sufficiently small. However, transfer to drinking water or locally produced food has little potential to lead to significant exposures to DU. Since poor solubility of uranium compounds and lack of information on speciation precludes the use of radioecological models for exposure assessment, biomonitoring has to be used for assessing exposed persons. Urine, feces, hair and nails record recent exposures to DU. With the exception of crews of military vehicles having been hit by DU penetrators, no body burdens above the range of values for natural uranium have been found. Therefore, observable health effects are not expected and residual cancer risk estimates have to be based on theoretical considerations. They appear to be very minor for all post-conflict situations, i.e. a fraction of those expected from natural radiation.

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1. Properties and occurrence of uranium

Uranium is a heavy, silvery-white, ductile and slightly paramagnetic metal, which is pyrophoric when finely divided. It is slightly softer than steel and reacts with cold water when present in a finely divided state. In air it easily oxidizes and becomes coated with a layer of oxide. Thus in nature uranium mainly occurs in oxidized form. Uranium is about as abundant as molybdenum and arsenic and more plentiful than mercury, antimony, tungsten and cadmium. It occurs in numerous minerals and is also found in lignite, monazite sands, phosphate rock and phosphate fertilizers. In ores it occurs as uranite (UO_2^{2+}), pitchblende ($\text{U}_3\text{O}_8^{2+}$) or as secondary minerals (complex oxides, silicates, phosphates, vanadates). Uranium is the heaviest naturally occurring element and is found at an average concentration of 0.0003% (3 mg/kg) in the earth's crust. In seawater the concentration is about 3.0 $\mu\text{g/l}$. Due to its presence in soil, rocks, surface and underground water, air, plants, and animals it occurs also in trace amounts in many foods and in drinking water. Table 1 shows typical concentration ranges in different environmental matrices.

The daily intake of uranium is estimated to be 1–2 μg in food and 1.5 μg in water consumed (ATSDR, 1999). The human body contains approximately 56 μg of uranium, 32 μg (56%) are in the skeleton, 11 μg in muscle tissue, 9 μg in fat, 2 μg in blood and less than 1 μg in lung, liver and kidneys (Fisenne et al., 1988). The uranium in the human body is derived mostly from uranium in food, especially from vegetables, cereals, and table salt (Priest, 2001; Fisenne et al., 1987).

2. Radiological properties of uranium

All isotopes of uranium are radioactive. Naturally occurring uranium contains three isotopes, namely ^{238}U , ^{235}U , and ^{234}U . ^{235}U and ^{238}U are the origin of two different decay chains. All uranium isotopes have the same chemical properties because they all have the same number of protons (92), but different radiological properties. The radioactivity of isotopes is dependent on the half-life. The most abundant naturally occurring uranium isotope, ^{238}U , has the longest half-life (Table 2) and, consequently, the lowest specific activity. Because of its higher specific activity ^{234}U contributes as much as ^{238}U to the radioactivity of natural uranium although the weight percentage of this isotope is extremely small (0.006%).

Table 1
Uranium values in environmental matrices

Matrix	Typical concentration range	Reference
Soil	0.3–11.7 mg/kg	UNSCEAR, 1993
Air	2.5×10^{-8} – 10^{-7} mg/m ³	NCRP, 1999
Surface water	3×10^{-2} –2.1 $\mu\text{g/l}$	WHO, 2001
Ground water	3×10^{-3} –2.0 $\mu\text{g/l}$	WHO, 2001

Table 2
Characteristics of uranium isotopes in natural uranium

Isotope	Half-life (years)	Relative mass (%)	Specific activity [Bq/g]
^{238}U	4.47×10^9	99.3	12,455
^{235}U	7.04×10^8	0.72	80,011
^{234}U	2.46×10^5	0.006	231×10^6

Natural uranium is considered a weakly radioactive element. In addition, uranium is categorized as a heavy metal with a chemotoxic potential (Burkart, 1988, 1991).

All natural uranium isotopes emit alpha particles, namely positively charged ions composed of two protons and two neutrons. Due to their relatively large size and charge, alpha particles rapidly lose their kinetic energy and have little penetrating power. The penetration range of a typical 5 MeV alpha particle is approximately 4 cm in air and only about 50 μm in soft tissue. Therefore they are unable to penetrate even the superficial keratin layer of human skin. As a result, uranium principally represents an internal radiation hazard. Uranium isotopes decay to other radioactive elements that eventually decay to stable lead isotopes. In the decay process, beta and gamma radiation is emitted. Beta particles have greater ability to penetrate the skin than alpha particles. Gamma rays are extremely penetrating and can present both an internal and external hazard. In nature, uranium is in secular equilibrium with the daughters of the decay chain. For this reason natural uranium together with its daughters yield four–five times as many decays per second as pure uranium. Radon (^{222}Rn), a radioactive noble gas in the decay chain of ^{238}U , can easily escape from soil or rock and, contrary to uranium, is a major contributor to the radiation exposure of the world population. When uranium is separated from its ores, the decay chain is broken. Only thorium (^{234}Th) and protactinium (^{234}Pa) reach equilibrium with ^{238}U within about 1 year and are the major contributors to the radioactivity of the purified uranium. The remaining members of the decay chain following ^{234}U take thousands of years to reach equilibrium and can be neglected. ^{235}U follows the same pattern and only thorium (^{231}Th) reaches equilibrium rapidly. The decay products of ^{238}U (^{234}Th and ^{234}Pa) and ^{234}U (^{231}Th) are responsible for the presence of beta and gamma radiation in purified natural uranium.

The major nuclear use of uranium is as fuel for nuclear reactors and in nuclear weapons. Of the uranium isotopes only ^{235}U (app. 0.72% of mass) is fissile. Consequently, during the manufacture of nuclear fuel for most types of reactors, the relative concentration of ^{235}U has to be increased. A byproduct of this enrichment process is depleted uranium (DU).

3. Depleted uranium (DU)

Depleted uranium (DU) is distinguished from natural uranium by lower relative concentrations of ^{235}U (<0.7%) and ^{234}U . In typical DU the content of ^{235}U is about

one-third of its original value (0.2–0.3%). Consequently, the activity of DU is about 60% the activity of natural uranium (see Table 3).

DU is 3 million times less radioactive than ^{226}Ra (still found in many old luminous clocks and watches) and 10 million times less radioactive than ^{241}Am , which is found in commercial fire detectors. Depleted uranium may contain traces of ^{236}U (<0.003%) from cross-contamination occurring when the same equipment has been used for handling both non-irradiated and irradiated uranium (TACOM, 2000). The long-lived uranium isotope ^{236}U ($T_{1/2}=23.48\times10^6$ a) is produced by ^{235}U neutron capture and builds up to high levels in nuclear fuel. This isotope can also be found in the natural environment as a result of nuclear activities, such as atmospheric bomb tests, waste dumping, the Chernobyl accident and the interaction of solar neutrons with soil uranium. DU penetrators collected in Kosovo were found to contain traces of ^{236}U and $^{239+240}\text{Pu}$ (UNEP, 2001). It has been also reported (Diehl, 2001) that trace amounts of Am, Np, and ^{99}Tc could also be present in DU.

Metallic uranium (including DU) is 65% more dense than lead (density of 19 g/cm³), has a high melting point (1132 °C), is highly pyrophoric, and has a tensile strength comparable to most steels. These properties, as well as the relative high availability and low cost, has led to various civilian and military applications of DU.

4. Applications of DU

4.1. Civilian applications

One intended use of depleted uranium was as a cladding material in fast-breeder reactors, where its interactions with neutrons would produce additional reactor fuel, in the form of ^{239}Pu . Depleted uranium was also used as a fluorescent additive in dental porcelain crowns (now discontinued), as X-ray radiation shielding in hospitals, as counterweights for rudders and flaps in commercial aircraft and fork lifts, and in the keels of sailing yachts. In the aircraft crash that occurred in Amsterdam on 4th October 1992, DU used as a counterweight, was released into the environment (Uijt de Haag et al. (2000).

Table 3
Comparison between the activity of natural uranium and DU (0.2% ^{235}U)

Isotope	Natural uranium Activity in 1 mg (Bq/mg)	Depleted uranium Activity in 1 mg (Bq/mg)
^{234}U	12.40	2.26
^{235}U	0.57	0.16
^{238}U	12.40	12.40
Total	25.28	14.80

4.2. Military applications

In the early 1970s, the US Army began researching the use of depleted uranium metal in kinetic energy penetrators and tank armor. High-density materials such as tungsten (density 19.3 g/cm³) and DU (density 19 g/cm³) were considered. DU was ultimately selected due to its availability, price and pyrophoricity (Danesi, 1990; Anderson et al., 1997). Tungsten has a much higher melting point (3410 °C) than uranium (1132 °C) and lacks pyrophoricity. Therefore, a tungsten projectile becomes blunt on impact and is less effective in piercing armor (Peterson, 1999). During processing DU penetrators are hardened by reducing the carbon content and by alloying with 0.75% by weight of titanium (Bukowski et al., 1993). The surface of a DU penetrator ignites on impact (especially with steel), due to the high temperature generated by the impact and the relatively low melting point of uranium (1132 °C). In addition, the projectile sharpens as it melts and pierces heavy armor (Rostker, 1998). DU projectile impacts are often characterized by a small, round entry hole (US-ACS, 1995). The 30-mm DU rounds, which were used by the US air force in the Gulf War and in Kosovo, can pierce steel armor up to a thickness of 9 cm. This DU 30-mm ammunition consists of a conical DU penetrator of 95 mm length, a diameter at the base of 16 mm and a weight of approximately 280 g. The penetrator is fixed in aluminum casing called 'jacket'. This has a diameter of 30 mm and a length of 60 mm. The A-10 aircraft is equipped with one gun firing 3900 rounds per minute. A typical burst of fire of 2–3 s involves 120–195 rounds. Normally the DU ammunition is present in about 75% of the rounds, the rest consists of non-DU ammunition. The shots hit the ground in a straight line and, depending on the angle of approach, hit the ground 1–3 m apart and cover an area of about 500 m². The number of penetrators hitting a target depends upon the type of target. In most cases not more than 10% of the penetrators hit the target (CHPPM, 2000). When the penetrator hits a hard object, e.g. an armored vehicle, the penetrator pierces the metal sheet, generally leaving the jacket behind. The DU dust which may be formed during impact can be dispersed and contaminate the environment. It is estimated that normally 10–35% (and a maximum of 70%) of the DU penetrator becomes an aerosol on impact or when the DU catches fire (Harley et al., 1999). Most of the dust particles have been reported to be smaller than 5 µm in size which keeps them airborne for an extended time, and will spread according to wind direction. DU dust is black and a target that has been hit by DU ammunition can be often recognized by the black dust cover in and around the target (US AEPI, 1995). After an attack with DU ammunition this will be deposited on the ground and other surfaces as partially oxidized DU fragments of different size, and as uranium oxide dust. According to investigations conducted at US test sites, most of the DU dust is deposited within a distance of 100 m of the hit target (CHPPM, 2000). Claims have also been made that the DU dust can travel up to 40 km and remain airborne for a considerable time (Belgrade, 2000). The majority of the penetrators that impact on soft targets (e.g. sand or clay) are expected to penetrate the ground to a depth of more than 50 cm, remaining intact for a longtime. Hits by depleted uranium on soft targets, e.g. non-armored vehicles or soil, do not generate significant dust contamination.

Depending on the chemical properties of different soils and rocks, the weathering of DU penetrators varies. In quartz sand, granite or acidic volcanic rock, solubilization rates may be high enough to lead to local contamination of groundwater. Wind and water will redistribute fine DU dust. Adsorption onto soil particles, mainly on clay particles and organic matter, will reduce mobility and the danger of resuspension. The major concern for the potential environmental effects by intact penetrators or large penetrator fragments is the potential contamination of ground water after weathering (UNEP, 2001; WHO, 2001).

The US army is also using depleted uranium as part of protective armor for tanks. When used in armor, DU is inserted into a metallic ‘sleeve’ in the regular steel armor of a tank.

5. Military use of DU

Many of the world’s armies possess or are thought to possess DU weapons (Harley et al., 1999). Depleted uranium weapons are regarded as conventional weapons. Ammunition containing DU was used in three recent conflicts:

1. 1991, Iraq and Kuwait (Gulf War);
2. 1995, Bosnia–Herzegovina; and
3. 1999, Kosovo.

5.1. Iraq and Kuwait

DU was extensively used by the US forces during the Gulf War. Apparently this is the only conflict where large DU projectiles were fired from tanks. The Air Force fired 783,514 rounds of 30 mm DU ammunition corresponding to 259 tons of DU (DoAF, 1997). The Army fired 9,552 DU tank rounds, corresponding to approximately 50 tons of DU and the Marine aviation expended DU ammunition corresponding to about 11 tons of DU. The United Kingdom is the only other country known to have fired DU munitions during this conflict. The UK Ministry of Defense estimates that its tanks fired less than one hundred 120 mm DU rounds, corresponding to about one ton of DU. The total adds up to 321 tons of DU.

5.2. Bosnia–Herzegovina

About 10,800 DU rounds (approximately 3 tons of DU) were fired during NATO air strikes in Bosnia–Herzegovina in 1994 and 1995, mainly around Sarajevo.

5.3. Kosovo

According to NATO information about 30,000 rounds of DU were fired from A-10 planes in Kosovo, corresponding to about 10 tons of DU. A total of 112 sites in and close to the border of Kosovo were hit with DU ammunition.

6. Analytical determination of DU

The determination of DU in environmental and biological samples requires the measurement of the concentrations of the two isotopes ^{238}U and ^{235}U . Based on the assumption that the ^{235}U content in DU is 0.2%, the ratio, R , $^{235}\text{U}/^{238}\text{U}$ in a typical environmental sample containing both natural uranium and DU can be calculated from Eq. (1), where X is the fraction of uranium present as DU and $1-X$ the part of uranium present as natural uranium.

$$R = \frac{0.72 - 0.52 \times X}{99.2745 + 0.5255 \times X} \quad (1)$$

The amount of DU is underestimated if the content of ^{235}U in DU is higher than 0.2%.

The concentrations of the different uranium isotopes can be accurately measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Thermal Ionization Mass Spectrometry (TIMS) or Secondary Ion Mass Spectrometry (SIMS). α - or γ -spectrometry have been also used, particularly in the past (Shoji et al., 2001; Baglan et al., 1999). However they have major limitations and disadvantages with respect to the mass spectrometric techniques both in terms of levels of detection, selectivity and rapidity.

Ejnik and co-workers (2000) developed a simple method based on ICP-MS to identify exposure to depleted uranium by measuring the isotopic composition of uranium in urine. A minimum concentration of 14 ng l^{-1} uranium is required to obtain a sufficiently stable ^{235}U signal to allow calculating an isotopic ratio. A method for discriminating between natural and depleted uranium was recently developed by measuring and analyzing the γ -ray spectra of some samples.

7. Exposure to DU

The exposure to DU is critically dependent on whether it is external or internal.

7.1. External exposure

Only the beta and gamma components of DU contribute to external dose. The affected organ is the skin. External exposure to DU mainly occurs during combat activities when DU aerosols are generated, or when DU fragments are picked up. The contact dose for macroscopic parts of DU is about 2 mSv/h and much less for dust. Soldiers in vehicles shielded with DU armor are externally exposed for longer time periods but at very low dose rates in the range of $1 \text{ }\mu\text{Sv/h}$.

7.2. Internal exposure

Internal exposure to DU can occur through three pathways:

1. Ingestion (food and water);

2. Inhalation (aerosol); and
3. Embedded fragments or contaminated wounds (mainly for soldiers).

DU can enter the body in the form of uranium metal from fragments and as uranium oxides from oxidized DU formed after impact on hard targets. The major uranium oxides generated are U_3O_8 , UO_2 and UO_3 (Harley et al., 1999). These three uranium oxides are relatively insoluble, dissolving only slowly in body fluids (weeks for UO_3 to years for U_3O_8 and UO_2). Uranium compounds are classified according to their solubility as type F (fast) [UF_6], M (medium) [UO_3] and type S (slow) [U_3O_8 and UO_2] (Table 4). The solubility of U_3O_8 lies between type M and S and this can also be classified as M-type. In the body fluids uranium is dissolved as uranyl ion (UO_2^{2+}), an ionic form that may react with biological molecules (Lin et al., 1993; McLean, 1995).

Uranium is absorbed into the blood, carried and retained in body tissues and organs. Once absorbed, uranium forms soluble complexes with bicarbonate, citrate, or proteins (Stevens et al., 1980; Cooper et al., 1982).

7.2.1. Ingestion

Ingestion of DU is not considered the major exposure pathway (WHO, 2001; UNEP, 2001). However, direct ingestion of contaminated soil must be taken into consideration, in particular for children, through hand contamination. Direct ingestion of contaminated soil by cattle and sheep as a pathway to humans has also to be considered. The uranium transfer factor from soil to pasture grass is low and has been reported to be equal to 0.036 (Tsukada and Nakamura, 1998). Since the contaminated areas are generally small, it can be assumed that food contamination will be negligible.

Uptake of uranium with drinking water is one of the major ways of incorporation of natural uranium (UNSCEAR, 2000). Contamination of drinking water with DU can be caused by DU dust, fragments or penetrators buried in soil (UNEP, 2001; EC, 2001). The contamination is strongly dependent on the speciation of DU, the acidity and the reducing properties of the contaminated soil and aquifer. US studies on test sites did not reveal any contamination of local ground waters, although high amounts of DU were dispersed to the ground (Morris and Meinhold, 1995). The

Table 4
Classification of uranium compounds according to their rate of solubility

Type F (fast)	Type M (medium)	Type S (slow)
Uranium hexafluoride (UF_6)	Uranium tetrafluoride (UF_4)	Uranium dioxide (UO_2)
Uranium tetrachloride (UCl_4)	Uranium trioxide (UO_3)	Triuranium octaoxide (U_3O_8)
Uranyl fluoride (UO_2F_2)		
Uranyl nitrate hexahydrate ($\text{UO}_2(\text{NO}_3)_2 \cdot 6 \text{H}_2\text{O}$)		

highest concentration was found in suspended sediment carried by run-off water (Becker and Varta, 1995).

7.2.2. Inhalation

Inhalation of dust is considered the major pathway for DU exposure both in combat and non-combat situations. There are two possible ways of generating DU aerosols.

7.2.2.1. Impact of penetrators on hard surfaces. The quantity of aerosol formed during impact depends largely on the hardness of the target. The heavier the armor, the more aerosol will form as the DU penetrator expends its kinetic energy piercing the armor. When DU hits lightly armored vehicles, round, golf ball-sized entrance and exit holes are made, penetrator degradation is limited and DU aerosolization is limited. Aerosol formation is enhanced if the penetrator breaks up and melts. Particles formed inside the vehicle either adhere to the inside surfaces or are released into the atmosphere through any opening. As they accumulate on interior surfaces, the particle size and mass may change due to agglomeration processes. Particle sizes vary greatly, from larger, easily visible fragments to very fine, respirable particles that can reach the lower parts of the lung. Special precaution should be given when entering contaminated vehicles. For persons present during impact or entering an armored vehicle shortly afterwards, the aerosols generated at impact may lead to considerable inhalation exposures. These impact aerosol levels will be much higher than those from resuspended DU particles arising from contaminated surfaces and venting to the outside. Resuspension is critically dependent on air turbulence inside a vehicle, surface conditions and physical activity inside the vehicle.

7.2.2.2. Fires. Studies have shown that in the absence of violent explosions, few of the particles generated during a fire are small enough to be caught up in thermal currents (Parkhurst et al., 1995). Most particles produced in fires inside a vehicle adhere to the interior walls, but openings can let particles escape into the surrounding atmosphere. Uranium oxides formed in fires have a low solubility, making the chemical toxicity to humans less important than the radiotoxicity. Exposure to DU aerosols generated by fires can also occur in non-military activities. For example on 4th October 1992 a cargo plane crashed into an apartment building near Amsterdam. The aircraft had been carrying depleted uranium as counterbalance weights and about 150 kg uranium had been found missing after the final clearance of the crash site. The radiation dose due to uranium oxide particles was estimated to be less than 1 μSv , with a worst-case scenario reaching an upper limit of less than 1 mSv (Uijt de Haag et al. (2000).

7.2.3. Wound contamination and embedded fragments

Wound contamination can occur during combat activities or later in the unlikely case of accidental bruising of skin on contaminated surfaces. In the latter case, wound cleaning will effectively decontaminate and the resulting exposure to DU can be expected to be negligible. However, embedded fragments not removable by surgical

means result in chronic, internal exposure. Sixty-two American soldiers, wounded with DU shrapnel when their tanks or armored vehicles were hit by friendly fire during the Gulf War, were studied for the effects of embedded fragments of DU shrapnel in their bodies (McClain et al., 2001). It was shown that the DU metal slowly solubilizes in the body fluids, and that several years after the war, blood and urine levels of uranium are elevated by up to two orders of magnitude (Hooper et al., 1999).

8. Monitoring of DU

The major risk for the general public of considerable exposure to DU is mainly due to airborne particulate matter generated when DU penetrators hit hard targets. Exposure assessment can be achieved by directly monitoring human beings or by monitoring the environment. DU in the lungs can be determined directly by lung counting (in-situ method). However, this technique is only applicable if the amount of DU in the lungs is more than a few milligrams. Therefore meaningful measurements can be only made in the case of extremely high exposure (Au et al., 1998). For exposures back in time, where only a small fraction of the amount initially inhaled remains in the lungs, as a consequence of the low specific activity of DU lung dosimetry cannot be applied. In these cases it is then more practical to monitor the environment, and to use individual monitoring only when environmental monitoring indicates significant levels of contamination and the potential of DU incorporation.

Air contamination and atmospheric deposition can be evaluated either by the direct collection of airborne particulate matters using air filters, or using suitable air pollution biomonitors. The first approach may provide quantitative surveys and information on the transport of pollutants. The second approach is a non-expensive and reliable means of air quality status assessment, giving also an indication of the past level of pollution (Smodiš and Parr, 1999).

In the summer of 1999, Kerekes et al. (2001) used air samplers located in the southern region of Hungary for assessing the amount of DU in air. The isotopic ratios revealed only isotopic ratios very close to those expected for natural uranium. However, they claimed that the slightly increased concentration of ^{238}U in particles under $2.5\text{ }\mu\text{m}$ of diameter was caused by dispersed DU dust emitted during the Kosovo war. This observation, although somewhat surprising, cannot be excluded by meteorological considerations and, if substantiated, would support a potential for long range transport of DU aerosols for extended periods after primary deposition.

Lichen and mosses are considered suitable biomonitors of atmospheric trace element deposition. Because they obtain most of their nutrition directly from the atmosphere, they are used in many parts of the world to assess and monitor air quality. (Thomas and Gates, 1999; Bennett and Wetmore, 2000; Böhm et al., 2001). Lichen morphology does not vary with the seasons, they usually live for long periods and accumulation of pollutants can occur throughout the year (Garty et al., 2001). Because they lack roots, epiphytic lichens do not have access to soil nutrient pools

and therefore have a high capacity to accumulate elements, including uranium, mainly by trapping atmospheric particulates (Haas et al., 1998; Delfanti et al., 1999; Caritat et al., 2001). However, there has been very little detailed work done in the field on how uranium is fixed by lichens. Uranium is accumulated in lichen talus under moist and dry conditions from airborne particles and dust and even tiny fragments of lichens may already contain readily detectable concentrations. In the recent international Kosovo field study lichens from barks of living trees, collected in areas close to 'target areas', showed the presence of DU, indicating earlier presence of airborne DU, even in areas where no widespread ground contamination could be detected (Sansone et al., 2001). This underlines the usefulness of using lichens as sensitive bioindicators in areas in which DU ammunition has been used. In locations where no lichen communities were found, superficial tree bark revealed identical results (UNEP, 2001; Bellis et al. 2001b). The composition of uranium in tree rings was recently used to study the uptake of DU (Jesse et al., 2001). The results show that uranium is mobile in oak trees and they are thus not effective long-term biomonitoring of uranium pollution. It is also worth reporting that tree barks have been used to distinguish between enriched and natural uranium released in the Tokaimura accident of 30 September 1999 (Bellis et al., 2001a).

The United Nations Environment Programme (UNEP) organized a fact finding mission to Kosovo in November 2000 and collected more than 300 samples (soil, vegetation, water). The UNEP team visited 11 sites out of the 112 attacked with DU ammunition during the Kosovo conflict. The results of the study were recently made public (UNEP, 2001). The general conclusions were that no widespread contamination of the ground surface by DU was detectable in Kosovo. Detectable ground surface contamination by DU was only measured in small areas a few meters away from where DU penetrators were found or localized near holes or dents in the ground caused by direct penetrator impacts. DU was found dispersed in the ground beneath the penetrators and jackets lying on the surface and sometimes was measurable to a depth of 10–20 cm. Since only a few penetrators were found, it was speculated that many penetrators have been already collected in clearing operations or are buried at a depth of some meters in softer ground. It cannot be excluded that in the long-term the latter scenario might contaminate local ground waters (UNEP, 2001). However the study concluded that no significant risk of air, water or plant contamination is expected in the short- and medium-term.

In May 2000 the ICRC invited 32 people working for the International Red Cross and Red Crescent Movement in western Kosovo to provide urine samples which were subsequently analyzed to determine the concentration of uranium. Several countries having peacekeeping troops in the Balkans were also surveying their soldiers (GSF, 2001; ITN, 2001). All results revealed normal levels of uranium, thereby indicating no substantial DU exposure among this group.

9. Exposure pathways and health effects

Until recently, the chemical and radiological toxicity of uranium and other actinides has been assessed mainly in view of occupational hygiene and natural occurrence

(for a review, see Burkart, 1988, 1991). Since its emergence on battle fields 10 years ago, DU has been repeatedly claimed to be the cause of cancer, leukemia and other health effects among troops, local populations and other personnel of humanitarian organizations, who were present at locations where DU ammunition had been used during conflicts (Gulf War, Kosovo, Bosnia, Montenegro). Although many reports (GSF, 2001; ITN, 2001; UNEP, 2001; WHO, 2001) have now shown that such claims are unfounded, it is worth summarizing the main health effects that external and internal exposure to DU may cause under different situations and assumptions.

9.1. External exposure

Health effects from external exposure are limited to skin contact. The dose rate to the skin in direct contact with a piece of pure DU is caused by beta particles and is about 2 mSv/h (US-AEPI, 1995). To exceed the current occupational dose limit for skin of 500 mSv/a, a DU object would have to stay in direct skin contact for more than 250 h. Nevertheless, direct skin contact with DU has to be prevented, both for the local population and for clean-up personnel. Since occupational exposure limits are set in a way to exclude unacceptable individual risks, skin lesion or cancer in this specific case, no visible health effects are expected from external radiation caused by DU left in the field.

Tank crews with DU shielding receive an additional whole body dose rate in the range of 1 μ Sv/h. This is close to the shielding effect of the tank from terrestrial and cosmic radiation. In view of the low occupancy factor for such vehicles, additional annual exposures are small in comparison with natural radiation doses, and potential health effects insignificant.

9.2. Internal exposure

Health effects related to internal exposure may result from either chemical or radiological toxicity.

9.3. Ingestion

For oral ingestion several uranium limits were established (Table 5).

All limits are based on effects observed in experiments with animals and include

Table 5
Existing limits for uranium ingestion

Reference	Tolerable daily intake (TDI) (μ g/(kg·d))	Annual limit on intake (ALI) (mg) ^a
ATSDR, 1999	1.0	25.6
Jacob et al., 1997	0.7	17.9
WHO, 1998	0.6	15.3

^a Based on 70 kg body weight

additional safety factors. For soluble uranium it is necessary to take into account its chemical toxicity. Current information indicates that 2–5% of ingested, soluble DU is absorbed into the blood from the intestines (Harley et al., 1999). The remaining 95–98% is eliminated rapidly. Only about 0.2% of ingested, insoluble DU is absorbed into the blood (ATSDR, 1999). The fraction absorbed into the blood is rapidly cleared, with approximately 90% leaving the body in urine within the first week after intake. The rest will be distributed to tissues and organs. In particular, about 10% deposits in the kidneys. Most of the uranium deposited in the kidneys is eliminated in a few weeks. About another 15% deposits in the bone. Uranium remains much longer in the bone compartment, to the extent that after 5 and 25 years, a few percent and about 1%, respectively, is still present.

The WHO limit for uranium in drinking water is 2 µg/l, a value considered to protect against possible renal effects (WHO, 1998). The limit established by the US EPA is 30 µg/l. Both limits are based on the assumption that a person of 70 kg body weight consumes 2 l water/day. However it must be added that the high natural concentration of uranium in drinking water found in some Nordic countries (highest median 34 µg/l, maximum value 750 µg/l) has not been found to affect the health of local population (Frengstad et al., 2000). Thus, it can be concluded that contamination of ground or drinking water by DU does not represent an acute risk. However, although in all realistic scenarios the doses and chemical toxicity would be negligible (UNEP, 2001, EC, 2001) monitoring of wells downstream from highly contaminated sites might be recommended as a precautionary measure (UNEP, 2001).

9.3.1. Inhalation

About 95% of inhaled particles greater than 10 µm aerodynamic equivalent diameter (AED) deposit in the upper respiratory tract (Harley et al., 1999). Most of these clear to the pharynx, and to the gastrointestinal tract. Particles below 10 µm AED can reach deeper pulmonary regions (bronchioles and alveoli) and deposit for considerable time. The amount of DU that is absorbed into the blood and deposited in tissue and organs depends mainly on particle size and the solubility of the uranium containing particle. This solubility determines how quickly and efficiently the body absorbs uranium from the lung. ‘Soluble’ chemical forms are absorbed within days while ‘insoluble’ forms generally take months to years. Toxic chemical effects are more likely to be associated with the more soluble forms of uranium while radiation effects are more likely to be associated with the insoluble forms, such as particles that are deposited in the lung and local lymph nodes and retained for extended periods of time. In blood, approximately 47% of the uranium forms a complex with bicarbonate, 32% binds to plasma proteins, and 20% binds to erythrocytes (Chevari and Likhner, 1968). Up to 90% of the dissolved uranium reaching the blood compartment is excreted within the first few days after a single exposure (Harley et al., 1999). The remaining 10% deposits in the bones, kidneys and other organs, from where it is mobilized over a longer period of time. According to an US study about 6.4% of inhaled soluble DU and 0.3% of inhaled insoluble DU is ultimately transferred to the kidneys (CHPPM, 2000). The kidneys are the critical organ for uranium chemotoxicity. For high acute exposures, precipitation of uranyl-carbonate complexes in

the proximal tubules is critical and may lead to impairment of kidney function which may lead to irreversible damage at very high exposures (ATSDR, 1999). Chelating compounds may be used to prevent or reduce kidney damage in such accidental situations (Harley et al., 1999).

In US soldiers with a high load of DU shrapnel, no indications of kidney disfunction were seen in tests made several years after the Gulf War. Since DU levels in urine are still about 100-fold increased as compared to controls or Kosovo veterans, effects on kidney function for the latter can be excluded.

Measurement of uranium excreted in urine is a sensitive method for determining the amount of DU inhaled. However, uncertainties in the assessed intake can be quite large because many assumptions concerning aerosol size, uranium solubility, and transfer rates between different body compartments must be made. Naturally-occurring uranium in food and water is an important confounder (Werner, 1997). Typically, an individual may excrete, depending on the dietary intake, between 0.01 and 0.4 μg of uranium in urine each day. To assess the intake of DU, it is therefore necessary to measure the isotopic ratio $\text{U}^{235}/\text{U}^{238}$. High resolution ICP-MS, having a detection limit for DU in urine of approximately 0.01 $\mu\text{g}/\text{day}$ is often used for this type of studies (Schramel et al., 1997; Ejnik et al., 2000).

The following table (Table 6) shows values for lung retention and daily urinary excretion rates (at different times after intake). The values have been calculated with recent ICRP models, and refer to an inhalation of 1 mg of DU from an aerosol.

With regard to chemotoxicity, the occupational exposure level is 0.2 mg/m^3 . It is unlikely that more than 25% of the DU is soluble or that dust to which people are exposed contains more than 10% DU. On this basis, 0.2 mg/m^3 of inhaled soluble uranium corresponds to 8 mg dust per cubic meter in post-conflict situation.

9.3.1.1. Radiation doses and risk. To keep radiation risk reasonably low, the annual dose limit for a member of the public is 1 mSv, while the corresponding limit for a radiation worker is 20 mSv (ICRP, 1990). The committed effective dose to an adult from the inhalation of insoluble DU is 0.12 mSv/mg. Thus, the annual limit of 1 mSv would be reached by inhaling about 8 mg of DU. Thus, an adult

Table 6

Excretion rates as a function of time since exposure and aerosol solubility; assuming an inhaled amount of 1mg of DU (ICRP, 1994)

Time	Moderately soluble aerosol		Insoluble aerosol	
	Lung burden (mg)	Daily urinary excretion rate (mg per day)	Lung burden (mg)	Daily urinary excretion rate (mg per day)
1 day	0.06	0.02	0.07	0.0007
1 week	0.05	0.0007	0.06	0.00002
1 month	0.04	0.0003	0.05	0.000008
1 year	0.004	0.00002	0.02	0.000002
10 years	Negligible	0.0000002	0.003	0.0000004

Table 7

Comparison of the inhalation limits for natural uranium and DU (ICRP, 1994)

	Dose factor (mSv/mg)	Annual Limit on Intake (mg) ^a	Air concentration (µg/m ³) ^b
Insoluble			
Depleted uranium	0.12	8.3	0.95
Natural uranium	0.22	4.5	0.58
Soluble			
Depleted uranium	0.0075	124	16
Natural uranium	0.013	75	9.4

^a Based on 1 mSv/a^b Based on 1 mSv/a, a breathing rate of 1 m³/h, and continuous exposure

breathing rate of about 1 m³/h translates into a limit for chronic exposure of 0.95 µg DU/m³. Table 7 compares values calculated with recent ICRP models with values for natural uranium.

The following table (Table 8) shows doses to an adult resulting from the inhalation of 0.5 mg of DU (0.2% ²³⁵U). These values have been calculated with ICRP models (ICRP, 1994), which assume an even distribution across the entire organ.

9.3.2. Embedded fragments

The clinical effects of DU exposure in Gulf War veterans having embedded DU shrapnel in their body were compared with non exposed Gulf War veterans (Hooper et al., 1999; McDiarmid et al., 2001). DU-exposed Gulf War veterans were found to excrete elevated levels of urinary uranium even 7 years after their first exposure (range 0.01–30.7 µg/g creatinine vs. 0.01–0.05 µg/g creatinine in non-exposed controls). The renal function parameters were found to be the same in the two groups, strongly suggesting that years of exposure to uranium at these levels do not impair kidney function (McDiarmid et al., 2000). The persistence of the elevated urine concentration of uranium suggests on-going mobilization from a storage depot which results in a chronic systemic exposure. However, although adverse effects on the kidney, the presumed target organ, were not detectable, neurocognitive examinations

Table 8

Values calculated for 0.5 mg DU intake using an ICRP model (ICRP, 1994)

Type	Effective dose (µSv)	Lung (µSv)	Bone surface (µSv)	Liver (µSv)
Type S	61	500	3.4	0.46
Type M	22	170	26	3.6
Type F	3.7	2.3	65	8.9
Natural Uranium Type M	460	940	18,000	610

demonstrated a statistical relationship between urine uranium levels and lowered performance on computerized tests assessing performance efficiency. According to the literature, the annual committed effective dose from embedded fragments can be estimated to 0.1 mSv (Phipps and Bailey, 2001).

Pellmar et al. (1999) and Miller et al. (1998) studied the effect of DU pellets implemented in rats. In this study the highest concentration of uranium in the kidney was found as early as 1 day after implementation. In addition several other tissues and organs showed significantly increased levels of uranium (spleen, liver, heart, lung, brain, lymph nodes, and testicles). This led to the conclusion that probably kidney and bones are primary reservoirs for the uranium redistribution from embedded fragments.

10. Cancer

The period between radiation exposure and an increased risk for cancer is generally quite long (ICRP, 1990). In adults, the median latency period may be about 8 years for leukemia and two- to three-times longer for solid cancers. Generally, the additional cancer risk for low dose exposure is assumed to be proportional to the radiation dose. It is difficult to detect an increased cancer risk due to radiation at doses lower than 100 mSv because the excess risk at low doses is small in comparison to spontaneous rates of cancers of the same type. Therefore, no direct experimental or epidemiological evidence can be obtained. In the absence of any direct evidence UNSCEAR (1993) has estimated the increase of cancer risk due to low radiation doses by linear extrapolation from highly exposed human populations, such as the survivors of Hiroshima and Nagasaki. These calculations show that the additional risk of a lethal cancer associated with a dose of 1 mSv is about 1 in 20,000. This represents only an insignificant increase in comparison with the normal cancer risk in Europe of about one in five, or 20%. These theoretical considerations also form the base of the assessment recently published by the World Health Organization (WHO, 2001) which also excluded any link between exposure to depleted uranium and the onset of congenital abnormalities or serious toxic chemical effects on organs. Mass screening of the population and the creation of an immediate, cleanup programme at the sites where depleted uranium was used in ammunition in the Balkans was therefore not recommended.

Well documented lung cancer risks in many cohorts of uranium miners (Harley et al., 1999), should not be used for the assessment of DU risks. The increased risk of lung cancer in uranium miners can be attributed to another group of radionuclides—radon and its short-lived decay products—in a synergistic interaction with tobacco smoke. Radiation exposures in miners from uranium proper are only a tiny fraction of those from radon and its short-lived decay products, and are still smaller than those from longer-lived members of the uranium decay chains such as radium, polonium and lead.

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