

## 6 Case studies and exposure scenarios

The aim of this chapter is to outline the factors which determine potential exposure routes for a range of receptors to DU. This is based on an understanding of potential linkages between sources of DU via pathways to environmental receptors, including humans. This chapter describes generic exposure scenarios that may be used to identify the principal exposure pathways for a range of environments and receptors.

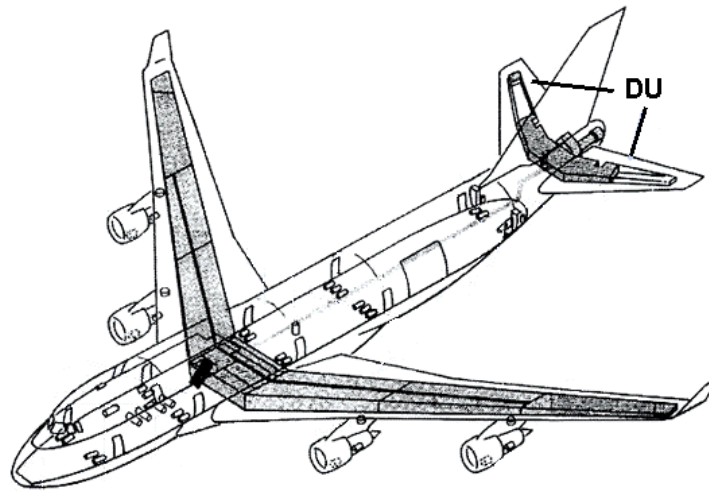
### 6.1 Case studies

#### 6.1.1 Potential exposure from air crashes

Under normal circumstances external radiation exposure of aircrew from intact DU balance weights during flight is insignificant when compared to the additional dose of ionizing radiation received because of exposure to cosmic rays during normal air travel (NUREG, 1999). However, workers may be exposed to DU used in aircraft during routine servicing of components in the immediate vicinity of any DU counterweights or to workers and members of the public as a result of an air crash. Levels of uranium introduced into the environment as a result of such an air crash depend on a wide variety of factors. To date, studies at the two most intensively investigated sites (Amsterdam and Stansted) have indicated that:

- whilst some DU may be released into the environment during air crashes, a large proportion of balance weights may be recovered from such crashes in a near intact condition.
- the varying quantities of DU balance weights used in aircraft, and the potential for production of small fragments and particles make it difficult to estimate the total percentage recovered or accounted for following an air crash.

As discussed in Chapter 4, DU is used as a material for balance weights (typically in rudders, elevators and ailerons) in some commercial aircraft (in weights of between 0.23 and 77 kg). While intact in the aircraft, the DU is either plated (cadmium and/or nickel) and painted or encased in a thin skin of aluminium alloy. Persons most likely to be exposed to DU in these conditions are those working in the manufacture of the components, in which case the conditions are as those discussed in Chapter 5, Section 5.3.5. Aircraft service personnel while unlikely to be directly exposed to DU may be exposed to beta and some weak gamma activity during service of components in the immediate vicinity of control surfaces and during replacement of counterweights (see also Chapter 9). The only environmental exposure route to DU from this source is when it is released as the result of an air crash, or when existing balance weights are inappropriately stored or scrapped. For example the presence of DU in a consignment of 53.5 metric tonnes of aluminium ingots sourced from 'recycled airplane parts' has recently been reported to have occurred ([www.nrc.gov/OPA/pn/pn301006.html](http://www.nrc.gov/OPA/pn/pn301006.html)).



**Figure 6.1** Schematic diagram of a Boeing-747 showing locations of DU counterweights (From US Federal Aviation Authority Document (FAA), CAS (2001)). Note this diagram does not acknowledge the use of DU counterweights in the Boeings wings that have been described elsewhere (e.g. NUREG, 1999).

Studies by Elder and Tinkle (1980), on the combustion of DU penetrators, concluded that oxide particles in the respirable size range could be formed when DU metal is exposed to temperatures greater than 500°C for burn times of longer than 30 minutes. Similarly, studies of the corrosion of metallic uranium in air and water vapour have established that corrosion was significantly more rapid in moist air (Haschke, 1998). While it may be that the casing or plating on DU counterweights protects it from fire and exposure to atmospheric corrosion, it is known that such protection degrades with time and that replating can be required. Results of burn tests and the questions raised in relation to the likely fate of DU in air accidents have also been highlighted in the media and in the scientific press (e.g. Parker, 1988, Uijt de Haag et al., 2000).

In the case of the 1992 Amsterdam plane crash (a wide bodied Boeing 747-258F), it has been reported in the press that only 130 kg of the initially estimated 282 kg of DU was recovered by clean-up teams, and that the Dutch commission of enquiry concluded that some of the 'missing' DU may have been released in the form of oxide particles (Kirby, 2000). Data presented in other recent studies such as Haag (2000) confirm that approximately 152 kg of the DU from the crashed plane remained unaccounted for almost eight years after the actual crash. This does not necessarily mean that this quantity entered the local environment, as some of this material could have been removed from the site during general clean-up operations that included the removal of significant quantities of top soil (Haag, 2000).

Using the limited data available Uijt de Haag et al. (2000) calculated potential radiological exposures *to bystanders* (for a 1 hour exposure) from the crash based on a 'worst case scenario' that all of the missing 152 kg of DU were oxidized into an insoluble respirable fraction, and a 'best estimate scenario' in which only 46 kg of the 152 kg were oxidized during the fire and that only 1% of this was in the respirable fraction. Modelling of airborne dispersion based on these cases resulted in atmospheric levels in the area in which bystanders were present of 3 µg/m<sup>3</sup> (best-estimate) and

2000  $\mu\text{g}/\text{m}^3$  (worst case estimate). From the results of their calculations they concluded that ‘it is therefore highly improbable that exposure of bystanders to uranium would result in the health complaints reported’.

In their calculations, Uijt de Haag et al. (2000) only considered exposure to bystanders present at the crash site for a relatively short period of time, that respirable uranium represented the greatest source of exposure to inhabitants because levels of uranium measured in soils were shown to be consistent with the natural background in the vicinity of the crash site and that 98% of any ingested uranium would be excreted within a few days. Such considerations would be less applicable for the assessment of longer term impacts where contaminated soils have not been removed from the site, although, obviously, levels of DU present in air and dust would have been significantly reduced due to fallout over the short to medium term.

### **6.1.2 Military uses**

The mobility and behaviour of uranium in the environment has been, and continues to be, extensively researched as a result of concerns related to the safe disposal of radioactive nuclear materials derived from power generation, from environmental pollution and regulations associated with the mining of uranium, and from studies assessing the exposure of populations to background levels of radioactivity and heavy metals. Despite this level of research, relatively few studies have been undertaken solely on the presence and environmental mobility of DU released into the environment through actual military conflict.

During the 1990s, the use of DU in test firings, military campaigns (i.e. operation Desert Storm, during the Gulf War, and operation Allied Force, during the Kosovo conflict) was the focus of much attention. This was principally, but not exclusively, due to a suggested or postulated link between exposure of military personnel, who had been in close contact with vehicles and installations that had either been attacked by armour-piercing munitions or had been protected by armour containing DU, and illnesses found in veterans from the Gulf War.

In the Gulf War it has been estimated that approximately 300 tons of DU was used sporadically over a total area in excess of 10 000  $\text{km}^2$  (Fetter and von Hippel, 1999) although another estimate based on data reported in CHPPM (2000) suggests that a total of 338 tons were used. The latter figure comprised 68 tons of large-calibre tank munitions, 260 tons of 30 mm armour-piercing munitions by US Air Force aviators and 11 tons of 25 mm armour-piercing munitions by US Marine aviators. During the air strikes in Kosovo, NATO fired about 10 tons of 30 mm armour-piercing munitions. NATO air operations in Bosnia–Herzegovina fired about 2 tons of 30 mm DU munitions.

The likelihood of exposure to DU and abundance of uranium in the environment following military activities was considered to be related to:

- the type of munitions used (for example single tank rounds with a high probability of impact (80% to 90%) versus strafing runs by ground-attack aircraft with a relatively lower probability of direct impacts).
- the density of munition use.

- the presence of aerosols and dust containing mixed oxides of DU.
- the presence of pieces of residual metallic DU.



**Figure 6.2** DU munition used by an A-10 Warthog aircraft and the Gatling gun from which it is fired.

It was considered during these studies that the pyrophoric nature of uranium was of special relevance to the potential health effects and environmental redistribution of uranium resulting from DU use in munitions and armour. Studies of the use of DU munitions have indicated that up to 70% of the DU in a given projectile may be converted to dust and aerosols on impact (AEPI, 1995). Other studies (CHPPM, 2000) indicate a lower estimate of 10% to 37%, depending upon the exact nature of the impact (i.e. with armour or other material such as concrete surrounding the target). This large discrepancy in reported conversion efficiencies may be due to variations in the hardness of the target, the velocity of impact and the angle of impact. The lower conversion figure agrees with information from the Pacific Northwest National Laboratory (formerly Batelle Pacific Northwest Laboratory) and indicates that when DU penetrators were heated under controlled environments (at about 1200°C) about 30% of the uranium was oxidized. In this case, over 99% of the formed uranium-oxide particles were greater than 20 µm AMAD (Mishima et al., 1985) and could therefore be considered as being non-respirable.

It has been observed that, in some cases, the DU projectile went completely through the target without oxidising or producing significant quantities of dust and aerosols, resulting in relatively large pieces of metallic DU entering the environment. Similarly it is likely that projectiles impacting into soft soil, surrounding hardened targets, may penetrate into the ground with minimal production of DU dust.

The percentage of such buried projectiles depends on engagement angles and ranges, soil types and terrain (AEPI, 1995) and is therefore extremely variable. For example during a typical strafing run against a single target, three planes may fire approximately 50 to 100 DU rounds each over an area of about 500 m<sup>2</sup> (10 meters wide by 50 meters long). In US airforce tests prior to the Gulf War a 'typical' A10 Thunderbolt strafing attack scenario against a T-62 tank resulted in a 90% miss and 10% hit rate (CHPPM,

2000). This indicates that a substantial mass of DU might become buried in a rural environment and lead to subsequent dispersion in the soil and leaching into groundwater as a result of chemical weathering. Little firm data appears to have been published on the potential penetration depth of projectiles into soils beyond observations that intact 30 mm and 25 mm penetrators have been found at a depth of 30 cm in soft soils typical of the Persian Gulf or Serbia (CHPPM, 2000). This is presumably because of the difficulty of detecting the beta or gamma radiation from buried DU projectiles. Projectiles that miss the target may also ricochet, skipping across the ground with minimal production of dust and aerosols. AEPI (1995) quote that such projectiles usually land within 2 to 4 km of the target.

During military conflict, particulate materials may also be produced during ammunition fires, such as may occur in ammunition depots or factories manufacturing and storing DU components. Elder and Tinkle (1980) have investigated the effects of simulated fires involving penetrators in storage or during transport. Experiments involved the initiation of semi-controlled conditions exposing the penetrators to high temperatures, an oxidising atmosphere and an intermediate wind speed of 2.23 m/s (5 mph). It was observed that penetrators did not tend towards self-sustained burning; this only occurs when finely divided uranium is oxidized. Depleted uranium aerosols were found to disperse in all forced draft oxidation experiments at temperatures in the range 500 to 1000°C. In an outdoor burning experiment with temperatures up to 1100°C, 42 to 47% of the penetrator by weight was oxidized in a three-hour burn. Outdoor burning also produced greater quantities of aerosols in the respirable range (<10 µm AMAD) with 62% of aerosol mass being in this size range compared to a maximum of 14% in the laboratory experiments. In general, DU aerosols in the respirable range are produced when penetrators are exposed to temperatures greater than 500°C for burn times of longer than 30 minutes. Some experiments have also indicated the presence of an ultra-fine particulate fraction (< 0.1 µm) often adhered to larger particles.

As a result of the high temperatures that are created during impact, uranium is converted to a series of oxides which include the relatively insoluble triuranium octaoxide (U<sub>3</sub>O<sub>8</sub>), uranium dioxide (UO<sub>2</sub>), and relatively soluble uranium trioxide (UO<sub>3</sub>) (Harley et al., 1999b). It has been stated that the relative insolubility of some of these oxides delays the rapid infiltration of dissolved uranium through the soil zone and into groundwater reserves. However, it does not preclude the physical migration and contamination of surface water resources with particulate uranium, or conversion into more, or less, soluble forms through interaction with other components of the target or soil (Patrick and Cornette, 1977). The exact chemical composition and crystalline structure of particulates and aerosols produced during the impact of DU projectiles also depend upon the composition of target material. For example, studies by Patrick and Cornette (1977) and summary text from CHPPM, (2000) indicate that complex spherical porous particles rich in iron and tungsten can be produced through high velocity collisions with armour. The same authors also state that similarly shaped, complex particles, may be formed by alloying with clay and sand (i.e. containing aluminium, potassium, silicon) as a result of direct impact with soil, or when hot, reactive, secondary particles from the initial impact interact with the soil environment.

Corrosion rates of any remaining metallic DU material in soils have been cited in AEPI (1995) to be in the order of 0.05 cm to 0.10 cm/year. Based on a 1 cm diameter by 15 cm long penetrator (e.g. about the same as a 30 mm round) this equates with the release of approximately 90 g/year. For the larger projectile typical of 120 mm munitions (3 cm×32 cm) this equates with a release of approximately 500 g of DU/year.

Based on these corrosion rates, the remains of such projectiles will only remain as metallic DU for between 5 and 10 years. In areas of low water infiltration it is therefore likely that the rate of migration of DU from the corroding projectile will be controlled by the solubility of secondary corrosion products (i.e. the high concentrations of uranium produced by corrosion in migrating fluids will exceed the solubility of many secondary uranium minerals, thereby promoting precipitation of secondary minerals rather than migration).

Concentrations of uranium in soils associated with the release of DU from military conflict are less well known and are likely to be less predictable than releases measured under more controlled 'proving ground' conditions. However, tests conducted by the US ballistics research laboratory, have shown that, although DU particles thrown into the air can travel downwind, the largest amounts of DU dust created on impact come to rest inside a penetrated vehicle, with significant amounts on the outside surface and within 10 m of the target (SAIC, 1990). Further information citing tests on hard targets at the Nellis Air Force Range in the US, indicates that DU dust from the impact of a 30 mm munition strike was deposited within 100 m of the target. Similar tests with a 120 mm penetrator that perforated a tank resulted in 90% of the airborne DU outside the tank remaining within 50 m of the tank. These dispersal patterns remained typical even after a fire began in a test tank and continued for over 12 hours (AEPI, 1995).

In their calculations based on data from available studies (shown below) CHPPM (2000) used the following data with respect to exposure to DU particulates in and around the immediate vicinity of, a target hit by a DU round:

- airborne Release Fraction 10%–37%.
- respirable Fraction 60%–96%.
- chemical Form  $\text{UO}_2$ ,  $\text{U}_3\text{O}_8$ ,  $\text{UO}_3$ .
- particle Size 1 to 10  $\mu\text{m}$ .
- solubility characteristics in Lung Fluid 1%–83% Class Y(S); 1%–20% Class W(M); 1%–43% Class D(F).

Note that the values in brackets refer to ICRP-66 (1994a) absorption types (Annex 4). Classes D,W and Y reflect retention half time of day (D), week (W) or year (Y) (ICRP-30, 1979). Also the more soluble a particulate the less radiological hazard and the greater the chemical hazard (see Chapters 7, 8 and 9).

A number of authors have used various theoretical scenarios to assess the likely hazards posed by the use of DU munitions in conflict (e.g. Fetter and von Hippel, 1999; Liolios, 2000; UNEP/UNCHS, 1999; CHPPM, 2000; SSI, 2000). Results of these studies indicate that the people at most risk of exposure to DU munitions are the occupants of vehicles actually attacked and penetrated by DU munitions. Members of the general population including those downwind of battlefields were not considered by these authors to be at risk of significant exposure provided that vehicles struck by DU munitions were made inaccessible to curious civilians (or soldiers).

These studies currently lack validation due to lack of data, and rely on relatively simplistic scenarios, complex modelling or low-resolution broad-scale modelling. However, later studies such as those undertaken by CHPPM (2000) use more recent data and realistic scenarios. As is the case with all such scenarios, they are subject to inaccuracies when considering site-specific issues that may enhance the potential of exposure to DU (i.e. the heavy use of DU munitions in close proximity to important localized water resources or areas of market gardens).

Whilst some authors have suggested that the use of DU munitions are unlikely to add significantly to environmental baseline levels of uranium in soils, it is important to consider that:

- uranium derived from the fragmentation of munitions may be more bioavailable, and possibly mobile, than residual uranium present in weathered soils (as for example demonstrated during investigations of soils contaminated by uranium from the Fernald site by Elless et al. (1997) and at military firing ranges by Becker and Vanta (1995).
- the relative importance of additional anthropogenically derived uranium is dependent upon the degree and rate of mixing, and the depth to which such material is incorporated and redistributed amongst the upper soil horizons.



**Figure 6.3** Kosovar Albanian children playing on military equipment following the Kosovo conflict. Such activities may lead to significantly increased probabilities of exposure to DU. (photo used with the permission of National Gulf War Resource Center, see [www.ngwrc.org](http://www.ngwrc.org) ).

For example if DU from the impact of a 4.85 kg penetrator (50% volatilized) were evenly dispersed over a radius of 10 m to a depth of 10 cm it would produce a uranium concentration in soils of approximately 96 mg/kg. This value is above that observed in most natural soils and similar to that observed in dusts in the Amman area of Jordan in which phosphorite has been mined (Smith et al., 1995). However, if a similar release of uranium was restricted to the upper 1 cm of soil, as might be expected from the deposition of atmospheric particulates onto uniform soils of a high clay content, then the resultant concentration, assuming even airborne dispersal, would be in excess of 960 mg/kg. While the presence of elevated concentrations in the near surface region of soil profiles is likely to reduce transfer to plants, it is more likely to facilitate inadvertent exposure to uranium in dusts and other re-suspended forms.

The most extensively researched releases of DU into the environment have occurred in areas used by the military to test munitions (proving grounds). For example, an investigation at the US Department of Energy's Los Alamos National Laboratory, conducted for the US Army, suggested that up to 100 metric tonnes of DU may have been expended. It was estimated that a small canyon with an area of 3.1 square miles had a DU inventory in the region of 35 000 kg (Becker and Vanta, 1995). Similar

quantities of DU were also used at military proving grounds in Yuma, Aberdeen and Jefferson in the USA (Ebinger et al., 1996; Ebinger and Oxenburg, 1997).

The United National Environment Programme in their post-conflict environmental assessment of DU in Kosovo (UNEP, 2001) concluded that:

- There was no detectable, widespread contamination of the ground surface by DU. The corresponding radiological and toxicological risks were considered insignificant or non-existent.
- Detectable ground surface contamination was limited to areas within a few metres of penetrators or penetrator impact.
- There was no significant risk related to these contamination points in terms of possible contamination of air, water or plants. The only risk would be through direct hand contact or the ingestion of contaminated soil. Based on reasonable assumptions on intake of soil the radiological risk would be insignificant while from a toxicological point of view the possible intake might exceed the health limit.

Although studies at such sites are useful for establishing the distribution of uranium immediately following dispersal, they provide little if any information about the longer term mobilisation and distribution of uranium. The most practical way to undertake such studies is to investigate the dispersal at natural sites of uranium mineralisation. A wide range of such 'analogue' studies have been undertaken in support of the nuclear waste disposal industry. They have clearly demonstrated that oxides of uranium, including uraninite and pitchblende ( $UO_2$ ), may be readily weathered (by oxidation and complexation with inorganic and organic ligands) and converted into more mobile soluble, forms of uranium that become incorporated into local surface waters, groundwaters, micro-organisms and plants (e.g. Basham et al., 1989; Hooker et al., 1989; Burns and Finch, 1999). Currently, there is a lack of comparison between data produced from these studies and that derived from DU alloys and associated particulates and aerosols. However, it should also be noted that weathering rates of particulate dusts produced during the combustion of DU weapons are likely to be enhanced over those of residual metallic uranium (Patrick and Cornette, 1977; Becker and Vanta, 1995) due to their inherently smaller particle size and correspondingly high specific surface area (i.e. area/unit mass of substance)

### **Military exposure**

Modelling of various exposure scenarios has been undertaken as part of environmental monitoring and decommissioning programmes carried out at US Army proving grounds that have become contaminated with DU. The Jefferson Proving Ground decommissioning programme modelled exposure scenarios, which have been documented in several published reports, (Oxenberg et al., 1999; Ebinger, 1998; Ebinger and Oxenburg, 1997; AEPI, 1995; Ebinger and Hansen, 1994). It should be noted that these studies only form examples of the results that may be obtained during case studies and should not be extrapolated to other sites, such as the Balkans and the Gulf, without careful consideration and justification. Three exposure scenarios were generally modelled in these studies in order to consider suitable uses for the site following decommissioning:

- i) An occasional user of the site visiting for 4 to 6 weeks of the year to hunt. All food and water would be brought onto the site by the user. Game animals would be consumed by the hunter.

- ii) A subsistence farmer consuming vegetables, dairy products and meat from crops and livestock produced on the site. Drinking water would be obtained from uncontaminated off-site sources. A fraction of the drinking water for livestock would be from contaminated groundwater, but the remainder would be from uncontaminated surface water.
- iii) As for scenario (ii) except that all drinking water would be obtained from contaminated groundwater.

This modelling exercise concluded that no risk to humans occurred from occasional use of the site, the largest exposure to DU in this scenario being from exposure to contaminated dust.

The farming scenarios showed some risk of exposure due to inhalation of contaminated dust, but by far the largest exposure resulted from the use of contaminated groundwater as drinking water, either by livestock or by humans. The overall conclusions of the modelling exercises were that subsistence farming presented a greater risk of DU exposure than did occasional use. However, in this particular study farming scenarios were not pursued in greater detail, because farming and permanent occupation were considered to be inappropriate end uses due to the presence of unexploded ordnance on both proving grounds.

There are, of course, many cases worldwide where exposure to mines has not prevented the continuation or resumption of farming activities. In such circumstances, it may be desirable to compare the potential risks associated with exposure to uranium with those associated with farming in close proximity to such obvious risks.

The work carried out by Ebinger et al. (1990; 1996) at the Aberdeen and Yuma proving grounds considered exposure to all components of the ecosystem. Depleted uranium was found in almost all samples and was present in most of the ecosystem compartments at Yuma (the semi-arid site) but not so much at Aberdeen. Measurable uranium concentrations were also found in aquatic endpoints (biota) at Yuma and in deer tissues at Aberdeen. Radiological effects were found to be insignificant at both sites, but there was some tentative evidence of toxicological effects. Erosion at Yuma was the primary mechanism of DU transport; wind deposition being of secondary and minor importance. At the wetter Aberdeen site, the main migration pathways were considered to be transport of suspended detritus in surface waters.

Concentrations of uranium in ecosystem components showed kidney content to be below threshold values in all species except for Kangaroo rats at Yuma in which histopathology indicated possible damage to kidney tissue (Ebinger et al., 1996). The consumption of dust, which had become adhered to foliage, was the most important exposure pathway for animals living in these sites.

Model projections of exposure over the next 1000 years at these sites (Ebinger et al., 1996; Ebinger and Oxenburg, 1997) indicate a gradual decline of the importance of particulate exposure together with a gradual increase in exposure to groundwater contamination over the next 100 years, before reaching a reasonably steady state condition between 100 and 1000 years (i.e. uranium particles become weathered releasing dissolved uranium into the water table or are physically removed from the area). Obviously such rates are extremely dependent on mineralogy of the source of uranium, local soil type and hydrological conditions.

Elless et al. (1997) and Elless and Lee (1998) undertook the characterisation of uranium contaminated soils at various US sites. Uranium was found to be associated with the silt and clay size fractions of soil samples analysed in these studies. In addition, mineralogical analysis indicated that the predominant form of uranium contaminant in these soils was an autunite-like phase (e.g. hydrated calcium U(VI) phosphate). In addition to this, major phase uranium minerals such as uraninite (UO<sub>2</sub>, U(IV) oxide) and coffinite (U(IV) silicate, USiO<sub>4</sub>) were also present (IV and VI refers to the ionic charge on the uranium atom). While uraninite and coffinite are generally considered to be insoluble (<0.01 mg/l), autunite the dominant mineral is only slightly soluble (0.1 to 0.2 mg/l) (Langmuir, 1978).

During these studies (Elless et al., 1997 and Elless and Lee, 1998) uranium solubility was determined before and after remedial treatment in support of performing a health-based risk assessment. Solubility of uranium was determined in carbonate-rich soils associated with the contaminated sites, and in background soils, using 75 and 300 day extraction tests performed with rain and groundwater. The results indicate the importance of anionic uranium carbonate complexes in controlling mobility, and that the major factor influencing uranium mobility was solubility control by primary mineralogical phases rather than sorption. The results also indicated that contamination of groundwater resources by DU derived from munitions was possible at the Fernald site, and that this contamination was promoted by the use of carbonate-based erosion control and road building materials.

Risk calculations and biokinetic modelling based on the resultant solubility measurements indicated that the risks were greatest from the soil ingestion pathway and the direct consumption of infiltrating groundwater. Interestingly, the lowest risk in this class was attributed to the inhalation of soil-derived dusts.

From the perspective of kidney toxicity, the greatest source of risk was derived from exposure due to the direct ingestion of infiltrating, contaminated groundwater. In all cases, the calculated level of risk was extremely sensitive to the solubility of uranium and it was recommended by the authors that this parameter must not be overlooked when assessing potential risks associated with exposure to uranium from the environment. It should be noted that whereas a 75 day extraction test may be applicable to the leaching of uranium during infiltration of rainwater, it is inappropriate in assessing solubility within the human gastrointestinal tract, where residence times are in the order of hours (Ruby et al., 1996). Similarly, the use of acid stomach simulants do not adequately account for dissolution of uranium in the neutral environment of the upper intestinal tract.

Short-term leach testing of residues from DU munitions at the Elgin test site, which had been used for test firing of DU munitions for over 20 years, indicated remobilization of uranium from soils and to a more limited extent in drainage sediments over a time scale of 0 to 20 days (Becker and Vanta, 1995). It was hypothesized by these authors that this comparatively rapid leaching of uranium was due to the abundance of small particles released from munitions during the combustion process (the majority of DU particles being associated with the fine clay and silt fractions despite the sandy nature of the soil). Analysis of cores showed transport of DU into the soil profile with baseline composition being reached at a depth of approximately 100 cm.

The longer term durability of relatively insoluble U(IV) oxides has been investigated during studies of the mobilisation of uranium dioxide stored in geological media with particular reference to the direct disposal of spent nuclear fuels (e.g. Cachoir et al.,

1996; Gallien et al., 1996). Under oxidising conditions, a two-step process was defined in the alteration mechanism.

- (i) Incorporation of oxygen and hydrogen correlated to a reduction in the volumetric uranium content (kinetic control).
- (ii) Formation and dissolution of schoepite ( $\text{UO}_3 \cdot 2\text{H}_2\text{O}$ ) (thermodynamic control).

Under reducing conditions, preliminary experimental results suggested an alternative mechanism. Gallien et al. (1996) measured the concentration of uranium under reducing conditions to be as low as  $10^{-11}$  molar. Other investigations, again undertaken during studies pertinent to the disposal of nuclear waste, have investigated the occurrence and weatherability of uranium oxides under natural conditions (so called 'natural analogue' studies). Such studies (e.g. Miller et al., 2000; Basham et al., 1989; Hooker et al., 1989) have shown that even reduced uranium oxides may become mobilized into ecosystems and the local environment over a period of tens, hundreds and thousands of years. These are time scales which are impracticable for studies in the laboratory and at proving grounds.

Case studies performed to date emphasize the wide variability in the behaviour of uranium and the comparatively small range of potential end-use scenarios that have been investigated. This is particularly relevant where uranium and/or DU has been released into an environment from which the exclusion of human beings is not a practicable option.

## **6.2 Environmental exposure scenarios**

Factors to be considered for assessing environmental exposures include variables related to soil composition and chemistry, climate, hydrogeology, land use and the mode and magnitude of exposure.

### **6.2.1 Soil**

A range of soil functions may influence, or be influenced by DU contamination (Note that in the context of this work contamination does not imply harm to any given endpoint; see glossary). These are:

- control of substance and energy cycles as compartment of ecosystems.
- basis for the life of plants, animals and humans.
- carrier of genetic reservoir.
- basis for the production of agricultural products.
- buffer inhibiting movement of water, contaminants or other agents into groundwater.

Scenarios associated with some of these functions are discussed in the sections that follow on plants, animals and groundwaters.

Clearly the potential for DU to contaminate soil depends upon the magnitude and nature of exposure. For example, large-bore munitions with a high probability of hitting hardened targets and fires involving DU munitions will potentially introduce more particulate DU and DU-oxides into soils than situations where small-bore DU munitions

have been used in strafing attacks. On the other hand, the amount of metallic DU introduced deeper into the soil profile is likely to be greater for typical strafing attacks, particularly in rural environments.

It is important to consider the influence of scale when considering scenarios. For example, it must be decided whether to consider a battle as a single diffuse source of contamination or as a series of point-source contamination incidents. Such a decision is site-specific, and to a certain extent depends on the proximity, distribution and sensitivity of various receptors.

Whilst the weathering rate of both DU-oxides and metallic DU is low, it is still a relatively rapid process compared with that of many natural soil minerals. However, the mobility of weathered DU in the soil profile is dependent upon sorption and mass transport properties of the soil (i.e.  $K_d$  and the water infiltration rate, see Chapter 3). Depleted uranium has been shown to be mobile in environments subject to high surface erosion and low infiltration rates, such as deserts. The variation in  $K_d$  of uranium with organic carbon content and soil pH (as described in Chapter 3) indicates that mobility is likely to be greater in semi-arid calcareous environments, or calcareous environments in which neutral to alkaline soil pH combines with a low organic carbon content. While mobility is greater in semi-arid calcareous soils, low net infiltration may significantly reduce the dispersal and mixing of DU.

Both point and diffuse sources of DU will weather and slowly become homogenized with uranium naturally present in the soil environment. Any increased weathering and mobility associated with specific forms of DU can be viewed favourably, due to the reduction of high levels of point-source contamination (i.e. dilute and disperse). Thus the exposure of receptors in the surface environment may be reduced. This implies that the level of contamination in a number of receptors, such as groundwater, and risks associated with any harmful, non-threshold effects, may be increased. Similarly, dispersal may significantly decrease the cost-effectiveness and technical feasibility of clean-up.

To date, difficulties in identifying DU penetrators that have missed their target and become embedded in the soil profile has limited the development of scenarios relating to the exposure of soil to DU from munitions. The collection of data on the depths to which various DU munitions penetrate in generic soil units would greatly assist any such developments and the design of suitable sampling strategies.

### **6.2.2 Water**

When firing of DU munitions occurs over land, DU contamination of water is likely to be dominated by transfer from direct soil deposition, due to the small surface area that freshwater generally covers. The transfer of uranium from the soil, or regolith, will be controlled by physical and chemical processes, which will in themselves be regulated by the climatic and geologic environment in which the contamination occurs.

The nature of DU entry onto the soil surface (e.g. fragmentation from impact with a target) or within the soil profile (e.g. near intact burial) will affect the rate and mode of transfer of uranium to the soil-water, surface-water and ground water environments. Fragmentation will increase the surface area, of any one projectile available to chemical and physical weathering. Small particles may be entrained in the near-ground atmosphere during dry (dusty) conditions. Overland water flow, from rainfall or snow thaw, will cause the physical movement of particulates to surface watercourses, and

ultimately into estuaries and near-shore environments. Physical translocation of particulate material into groundwater may occur through the regolith and within aquifers that have secondary fracture flow mechanisms. The entire burial of DU weapons, from a 'soft' impact with soil, will lead to little fragmentation, but potentially contaminate groundwater resources through dissolution and migration into aquifers.

The mobility of DU in the near-surface environment will be controlled by factors such as the pH of soil minerals and water, and the sorption potential of soil minerals (Chapter-3). Thus where soil strongly binds the uranium in secondary phases or on surfaces (e.g. iron oxides, clay minerals or organic carbon), its release into soil water, and translocation to groundwater, should be minimal. In deeper environments mobility and attenuation are controlled by the composition of fracture coatings and water chemistry. Where uranium is highly mobile, water resources may be more vulnerable to contamination.

The vulnerability of water to uranium contamination will be controlled by the geological, soil conditions and mobility encountered. The primary factors affecting vulnerability, assuming that uranium is mobile, are the depth of the unsaturated zone (i.e. proximity of the contamination to the water table) and the infiltration rate of recharge. For example, vulnerability of water resources hosted in river gravels may be high due their proximity to the surface. Whilst, the vulnerability of those obtained from deeper, possibly confined, aquifers will be lower.

### **6.2.3 Plants**

Plants, while generally considered to be poor accumulators of uranium, may be affected by the presence of elevated uranium concentrations in the environment. Exposure to uranium can occur through water (soil pore water, irrigation water or rainfall), from the atmosphere (i.e. wet and dry deposition to foliage), from leaching of soil by root exudates or from the direct incorporation of particulate matter. During studies at the Yuma proving ground, the association of uranium and DU with vegetation and plant litter was clearly established, although the majority of determined uranium was considered to be associated with particulate contamination of samples that was easily removed by washing.

Any scenario exposure to plants occurs primarily through:

- 1) Atmospheric fallout of particulate material. This mode of contamination would therefore be more likely where conversion efficiencies of metallic DU into particulates and aerosols are high (i.e. tank attack onto armoured targets) or following an intense fire in which DU is present. There is clear evidence that such contamination may be minimized through thorough washing of vegetables, greens and fruit.
- 2) Uptake of contaminated water. The mobility of uranium in the soil zone (see 6.2.1 above) and the proximity of uranium to the root zones of plants principally control this mode of contamination. The relative importance of infiltrating uranium being carried in from the dissolution of surface particulates compared to that derived from the weathering of buried DU penetrators is therefore dependent upon the depth of penetration. Although uptake into the plant may be minimal, it is possible that uranium may become concentrated into the skin of vegetables and tubers during the exclusion process (this has been demonstrated in some non-food plants; e.g. Basham et al., 1989). Exposure may therefore again be reduced by careful peeling and washing.

#### **6.2.4 Animals**

Animals (domestic or undomesticated) may become exposed to DU through similar routes to humans. Domestic animals are more likely to be given fodder grown outside the affected area than wild stock, lessening their exposure. Animal exposure is likely to be greater than that of humans, due to their often monotonous and less spatially diverse diet. Herbivores typically ingest considerable quantities of soil while browsing and may be particularly vulnerable to particulate DU present in soils and adhered to vegetation surfaces. Carnivores may be affected by bio-accumulation from species lower down the food chain.

### **6.3 Human exposure scenarios**

When considering human exposure to uranium in the environment, it is important to consider the relative importance of the various exposure routes outlined above and in the previous chapter. An awareness of the most significant exposure pathway in any given circumstance allows the prioritisation of hazard assessment and control. For example, if it is recognized that the most important exposure route in a given scenario is posed by the inhalation of dust, immediate measures can be taken to prevent inhalation by suitable respiratory protection, exclusion from the area or by minimising wind-blown dust through irrigation. If, however, the greatest probability of exposure is posed by ingestion of contaminated food, measures can be taken to obtain alternative food sources.

The first six scenarios consider exposure routes associated with the use of DU munitions. Scenarios 7 and 8 represent exposure as a result of an air accident involving an aeroplane carrying DU counterweights. Scenario 9 considers exposure in the controlled environments of the uranium mining and processing industries.

#### *Scenario 1: Person in attack zone*

A person is in an area where the active use of DU munitions is occurring. The potential for dermal contact is high and this may even take the form of wounding with fragments of DU. The likelihood of exposure to DU aerosols and fine particulates as the result of impacts is high. Amounts of ingestion are considered low and would be principally associated with particles adhered to food.

#### *Scenario 2: Relief worker in a war zone*

A relief worker is entering a war zone to attend injured persons and assist with the clearing of associated debris. The exposure potential is similar to that in Scenario 1; the difference is that dermal contact is slightly reduced, and there is no risk posed by flying fragments of DU munitions or armour. There is likely to be greater contact with dust and soil, as people and debris are moved as part of the relief work. It has been suggested that levels of DU in re-suspended dusts are much lower than those encountered immediately after a high-energy impact as in Scenario 1 (CHPPM, 2000).

#### *Scenario 3: Metal reclamation*

Damaged armoured vehicles are decommissioned after military operations. Vehicles have often been transported and stood in the open air for periods of time. Unused munitions are removed, and in some cases vehicle parts may be salvaged for re-use. It is also possible that local inhabitants may take metal parts for their own use. This may

include armour containing DU. In this scenario welding and cutting equipment may be used (note this exposure scenario has been quoted in AEPI (1995) as potentially producing unacceptable levels of particulate DU). Dermal contact directly with DU metal and dust represents a potentially significant mode of exposure. Secondary mobilisation of dust may lead to inhalation, and welding, and cutting activities may result in the formation of fumes or aerosols, which may also be inhaled.

#### *Scenario 4: Local inhabitant*

Following the cessation of military activities in an area, subsistence and commercial farming may recommence. A farmer/plot holder and family will cultivate the soil and consequently be exposed to fine particles contaminated with uranium that may be inhaled or ingested. Contaminated soils may also be ingested as a result of hand to mouth activities. There is some potential of dermal exposure to metal fragments and dust. A subsistence farmer and his family will also rely on food and water that may be contaminated with uranium. The time scale of this will vary and be dependent on the bioavailability and mobility of the uranium in the environment. This will need to be assessed on a site-specific or regionally specific basis. Current estimates indicate that DU munitions and armour degrade and chemically weather over a period of 100 to 1000-years (Erickson, 1990), although this is highly dependent on the particle size or composition of any metallic uranium or uranium alloy; conceivably in some environments this could be considerably less. Certain traditional farming methods involve close contact with the soil and a stooping position that may encourage the inhalation of dust. Other more mechanized methods may result in the enhanced mobilisation and thus exposure by inhalation. Potential exposure to young children accompanying adults may be high.

An inhabitant in an urban area where a plane crash has occurred or has been subjected to bombardment may obtain some food from a vegetable patch or allotment garden tended by themselves. The key differences from a rural situation are that drinking water and dairy products are likely to be obtained from uncontaminated sources.

#### *Scenario 5: Children playing*

The return to normal activities in an area where DU munitions have been deployed will include children playing. This may be in areas where derelict military equipment remains. The hand to mouth and inquisitive activities of children may lead to significant dermal contact with metal fragments and dust. Ingestion of contaminated dust and soil will be likely and ingestion of contaminated food and water may also occur. Secondary mobilisation of fine contaminated particles may also increase potential exposure from inhalation. This scenario will be of much greater importance in regions in which geophagia is practised.

#### *Scenario 6: Villager*

Again this considers exposure following the cessation of military activities. People returning to villages in areas which have been affected by military activities may be exposed to DU in this environment (e.g. through the resuspension of dust or presence of metallic penetrators). The most important exposure routes are those related to the ingestion of drinking water and soil and dust, which are likely to be of greater importance than foodstuffs. It is likely that in most cases foodstuffs will be derived from a variety of local sources (or possibly from even wider afield). Drinking water and soil

are probably more consistent factors controlling exposure although contamination of drinking water supplies will vary according to source and method of supply etc.

*Scenario 7: Relief worker at an aircraft accident*

A relief or salvage worker, or possibly an investigation team, will work in the area in and around an aircraft crash site. Exposure routes are generally similar to those associated with relief workers in war zones. The magnitude of exposure is likely to be reduced because of the nature of the DU involved. Counterweights are large pieces of encased metal, whereas munitions consist of uncased metal which is designed to impact, and penetrate tank armour. Therefore, volatilisation and fragmentation are less likely and the relative importance of associated exposure pathways should be reduced. In the case of food and water ingestion, relief workers will be obtaining sustenance from external sources, and eating and drinking are likely to take place in areas removed from the crash site.

*Scenario 8: Local inhabitant near a plane crash*

An inhabitant of an area where a plane crash has occurred is assumed to obtain some food from a vegetable patch or allotment garden tended by themselves. This is a similar scenario to that of a subsistence farmer in a war zone. The key differences are that in the air crash zone drinking water and dairy products are likely to be obtained from uncontaminated sources.

*Scenario 9: Industrial exposure*

The potential exposure of workers would be expected to be controlled through national legislation and international recommendations which are designed to limit exposure. The greatest potential source of intake is likely to be by the inhalation of dust and fumes, although uptake through contaminated wounds, ingestion and dermal absorption cannot be excluded. Any contamination of food and drinking water will result from the inadvertent presence of dust while at work; or alternatively from off-site transport of dusts or waste products to the household environment. Because of the specific nature of these industrial processes, appropriate monitoring of the working environment and of the individual may be necessary. A guide on the assessment of exposure to uranium from mining and associated activities is available from the IAEA (1989a) which, despite ignoring the chemical toxicity of uranium, represents a useful approach in calculating and minimising exposure. Advice on individual monitoring procedures is given in ICRP-78 (1997).

*Scenario 10: Background exposure*

Background exposure illustrates the relative potential importance of various exposure pathways in an area unaffected by man-made sources of uranium or DU.

**Scenario summary** The above scenarios give an indication of the most likely exposure routes in a given situation. However, the specific conditions in an actual situation may have a major influence on these assessments and must be considered. For example, the potential magnitude of exposure posed by inhalation and ingestion of dust and aerosols in scenarios 1, 2 and 7 above would be greatly reduced if the events took place at a time of heavy rain. All exposure assessment and subsequent risk assessment exercises require the consideration of material and site-specific factors.

There are several key factors that can be noted from the scenarios:

- Exposure is likely to be greater under uncontrolled conditions.
- The potential for exposure during military conflict in which DU munitions and armour is used is more significant than that posed by the release of DU through air accidents.
- The relative importance of a variety of exposure routes in a war zone situation is similar for a range of scenarios. The actual health risk may however be of a very different magnitude, depending on concentrations in air, water etc. and pattern of exposure (e.g. acute or chronic).
- Outside of direct exposure to DU in active warfare, those with a large potential for exposure to DU are children returning to normal activities within a war zone. The hand to mouth action and inquisitive play of children mean that they are the most likely to be exposed in a wide variety of exposure pathways.

## 6.4 Summary

Humans and animals may become exposed to uranium by inhalation, ingestion, dermal sorption or injury (e.g. embedded fragments). The relative importance of each of these exposure routes depends on the physical and chemical nature of the uranium to which individuals may be exposed. For example exposure to uranium in a typical baseline environment away from anthropogenic or geological sources is likely to be dominated by ingestion of drinking water. In some cases, exposure may be dominated by the deliberate consumption of soil where geophagy is practised. Alternatively, for coastal populations, exposure may be dominated by the ingestion of shellfish. Estimates of baseline exposure to uranium range from 0.0005 to 0.001 mg/day.

The pyrophoric nature of uranium means that when DU munitions are used in warfare or training, a quantity of dust composed of mixed uranium oxides may be liberated in the immediate vicinity of any impact. Such dust may be inhaled, or enter the food chain either directly through the ingestion of dust on food or by plant uptake from surface water, via the deliberate or inadvertent ingestion of soil by young children, via the use of surface water for drinking or following migration of uranium into local groundwaters which may be used for drinking water or crop irrigation. While the ingestion of foods or soil and dust are likely to lead to exposure immediately following a period of conflict incorporation and migration to groundwater may occur over a time span of tens, hundreds of thousands of years.

During or after a military conflict exposure is also likely to be controlled by the type of engagement and the type of DU munitions used. For example strafing attacks may produce markedly different exposures than those likely to occur as a result of tank battles.

Due to the wide variation in potential exposure scenarios to uranium from natural and anthropogenic sources, it is recommended that exposure assessments are performed in a tiered manner (Tier 1: desk assessment and review; Tier 2: field study and analysis; Tier 3: detailed site-specific exposure assessment/validation) prior to any decisions being

made on the use of remedial measures or likelihood of potential health outcomes.

Current gaps in knowledge include:

- Weathering of DU combustion products and bioavailability (long-term and short-term, appropriate climatic scenarios).
- Relative importance of soil ingestion (regional dependence).
- Validation of actual exposure (natural and following conflict).
- Ecological cycling (food stuffs and plant uptake).

## **7 Behaviour of uranium in the body**

### **7.1 Introduction**

Although ubiquitous in the environment, uranium has no known metabolic function in humans and animals and has been generally regarded as a nonessential element. However, as in the case of many heavy metals, the search for health effects related to short-term and long-term exposure to uranium has an extended history. The first recorded reference to studies on uranium exposure dates back to 1824, only 40 years after its discovery. Later in the same century, studies on human subjects showed that uranium could be administered as a therapeutic agent for diabetes as it had been shown to increase glucose excretion (Hodge et al., 1973). During the final decades of the 20<sup>th</sup>-century, research into the occurrence and toxicity of natural uranium continued to be undertaken, with increasing emphasis being placed on understanding natural and baseline exposure to uranium and uranium-series radionuclides, on the toxicity of short-lived decay products such as radon and its progeny, and on the effects of chronic long-term exposure to natural uranium. These developments occurred in parallel with the development of increasingly sensitive and robust analytical techniques for the determination of uranium in soil, water and stream sediments primarily developed for application in the exploration and exploitation of uranium resources. Such techniques have also proved of great value in improving our understanding of how uranium behaves in the body.

As has been indicated in numerous scientific and general interest publications over the past decade, very little health-related research has been undertaken specifically on DU. The major reason for this is the extensive research undertaken on natural and enriched uranium, both of which pose a greater radiological hazard and an identical toxicological chemical hazard to depleted uranium. However, it is also important to realize that any resultant health effects from exposure to depleted uranium, due to radioactive decay or chemical interactions, do not necessarily occur in isolation. For example, although no direct information appears to be available on this subject, ATSDR (1999) points out that co-exposure to other nephrotoxins (e.g. lead and cadmium) could have an additive effect. Similarly, little reliable data exists on synergistic effects that may lead to enhanced uptake or excretion of uranium compounds.

### **7.2 Biodistribution and toxicokinetics**

Although uranium has no known metabolic function, its strong affinity for many physiological compounds suggest that it is unlikely to exist, except transiently, as free ions. For example, given the near-neutral pH of blood and many body fluids, it is generally accepted that the most important species controlling uranium mobility in the systemic circulation are the carbonate, bicarbonate species and citrate complexes of U(VI) (Cooper et al., 1982). For example, about half of the U(VI) circulating in blood is present as carbonate complexes (e.g.  $\text{UO}_2\text{CO}_3$  and  $\text{UO}_2(\text{CO}_3)_2^{2-}$ ) (Durbin, 1984) or associated with bicarbonate and citrate complexes (Cooper et al., 1982). However, at slightly lower pH, in the range 5 to 6, uranium is also known to complex strongly with a wide range of organic ligands and these might be expected to impact on the assimilation of uranium into body tissues from the systemic circulation. In urine, uranium is present predominantly as the bicarbonate complex (Cooper et al., 1982)

Uranium has been shown to form strong complexes *in-vitro* with biological molecules containing phosphate (e.g. glucose phosphate, phospholipids and nucleic acids and for glucosamine, acetylglucosamine and related polymers) (Guibal et al., 1996; Wedeen, 1992), although the presence of some of these compounds has yet to be established *in-vivo*.

The ICRP biokinetic models for predicting the behaviour of uranium in the body are described in more detail in Annex 4. Other toxico-kinetic models covering the systemic behaviour of uranium include those proposed by Sontag (1986), Fisher et al. (1991) and Wrenn et al. (1988).

### 7.3 Ingestion

Absorption of uranium from the gastrointestinal tract (GIT) depends on the bio-solubility of the uranium compound, previous food consumption and concomitant exposure to oxidising agents. Wrenn et al. (1985) quoted a value of 1% to 2% average human gastrointestinal absorption. This has gained general acceptance and the currently recommended generic average human gastrointestinal absorption value for uranium is considered to be about 2% (WHO, 1998b; Leggett and Harrison, 1995; ICRP-72, 1996). Studies of GIT uptake factors in animals showed variance with quantities of uranium administered, age, dietary stress such as iron deficiency, and fasting. For example, various studies cited in ATSDR (1990) indicate factors of 3 to 4 times enhancement under these conditions compared to values obtained during studies on well-nourished adult animals. Uptake factors for specific modes of ingestion are discussed below.

*Drinking Water:* Uranium dissolved in water is almost exclusively present as the hexavalent species, and hence uptake factors for this form should generally be applied during the calculation of uptake from the human gut. For example the IAEA (1989a) suggested a gut uptake factor for uranium in water of 5% for most hexavalent compounds and a factor of 0.2 % for  $\text{UO}_2$ ,  $\text{U}_3\text{O}_8$  and most tetravalent compounds. More recent data from Harduin et al. (1994) suggest that 5% may be a conservative estimate and that a lower uptake factor of 2% for soluble components in water should be adopted.

*Food:* Wrenn et al. (1985) reviewed uranium uptake factors across a range of average diets and suggested an uptake factor of 2% to 3%. However, these authors report that values as high as 20 % have been quoted in the literature. It should be noted that these authors also report a significantly greater uptake factor in rats than other species, and suggest that rats should not be used to determine uptake factors for man.

*Soil:* The bioavailability of uranium in soils depends on the physical–chemical form in which the uranium is present. It is therefore impossible to suggest a single value or range of values associated with soils, particularly as the diversity of potential chemical forms of uranium and soil types is large. Characterisation and solubility measurements of uranium contaminated soils undertaken by Elless et al., (1997) confirm these general points and emphasize the wide variability in solubility (and inferred bioavailability) of uranium from various sources. However, given our current knowledge of conditions in the human gut and the aqueous chemistry and mineralogy of U, it is reasonable to hypothesize that uranium mobility (and thence bioavailability) is likely to be higher in the neutral conditions of the upper intestine, than in the acid conditions of the gut. This may be influenced by the formation of stable oxy-anion complexes. Consequently, the use of acidic leachates to assess solubility as described in Elless (1997) may yield

different results from those obtained using other forms of physiologically based extraction tests (e.g. Ruby et al., 1996)

## 7.4 Inhalation

Clinical postmortem studies of an occupationally exposed worker indicate that significant amounts of uranium are present in lung tissues (Kathren et al., 1989). This indicates inhalation has been an important source of accumulation.

The amount of particulate material deposited in the respiratory tract will depend on several factors, which include particle size and shape, breathing rates etc. These factors have been described in detail in the ICRP Human Respiratory Tract Model (HRTM) (ICRP-66, 1994a), see also Annex 4. Absorption of inhaled uranium into the systemic circulation will depend on the rate at which particles dissolve in the lungs and on their interactions with the ligands present in lung fluid.

Generic absorption parameters for soluble (Type F), moderately soluble (Type M) and poorly soluble (Type S) compounds have been published by ICRP. These are also summarized in Annex 4, along with factors influencing lung retention.

**Table 7.1** Absorption types for uranium compounds (ICRP-71, 1995b).

Type	Typical Compounds
F	UF <sub>6</sub> , UO <sub>2</sub> F, UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub>
M	UO <sub>3</sub> , UF <sub>4</sub> , UCl <sub>4</sub> , U <sub>3</sub> O <sub>8</sub>
S	UO <sub>2</sub> ,

F Fast; M Moderate; S Slow

Note some preparations of U<sub>3</sub>O<sub>8</sub> can be assigned to Type M while for others their behaviour lies between Type M and Type S.

The behaviour of all the compounds formed in the nuclear fuel cycle has been studied extensively. Recently, much of this work has been reviewed and material-specific values have been reported in terms of HRTM parameters (Hodgson et al., 2000; Ansoberlo et al., 2001). When sound experimental data are available ICRP has recommend for many years that these should be used instead of the default given in the HRTM model. Material-specific biokinetics and toxico-kinetics of uranium oxides are discussed further in Chapters 10 and 12.

In the absence of material-specific *in-vivo* data, *in-vitro* dissolution studies (Edison, 1994) can provide much useful information which can be of use in assessing the implications to exposure.

It is recognized that there is a paucity of data on the dissolution and absorption characteristics of aerosols formed as a consequence of the combustion and thermal oxidation of DU. In this context, *in-vitro* studies and an improvement in our understanding of the mineralogical nature of these particles will provide rapid and cost-effective information given the physical-chemical variability of such materials resulting from different scenarios and outcomes.

*In-vitro* solubility analysis of particles (< 10 µm AED in diameter) produced immediately following the impact of DU munitions found that between 24% and 43% of the total particulate load dissolved rapidly. The remainder of the particles were relatively insoluble with a predicted lung retention half time of longer than 100 days (Jette, 1990). While these data may be used to estimate the radiological dose received from inhalation of such particles immediately following impact, further study is required to assess the impact of weathering on particle solubility class.

## **7.5 Injury, insult and dermal sorption**

Increased concentrations of urinary uranium (up to 150 times compared to those observed in control groups without fragments) have been observed in soldiers with retained DU fragments (Hooper et al., 1999). These studies also revealed a slow continuing release of uranium from individuals with fragments over one year later, suggesting a slow controlled release of DU. Parallel studies undertaken on rats have reported increased concentrations of DU in the kidneys and bone, although detectable amounts were also observed in the brain, testicles and lymph nodes of exposed animals (Pellmar et al., 1999a) (see Chapter 8 for further details). Further validation and research into the implications of this data are required.

It has been demonstrated in animal studies that soluble uranium compounds such as nitrate can be absorbed through the skin (Orcutt, 1949; DeRey et al., 1983, 1984). In studies with rabbits, death due to renal failure was observed to occur via this mode of exposure with a lowest LD<sub>50</sub> value of 28 mg uranium per kg as uranyl nitrate in an ethereal solution (Orcutt, 1949). Rats and guinea pigs were observed to be significantly less sensitive. More recent studies of sub-acute dermal exposure to uranyl nitrate (typical applied concentrations 0.6 g/ml uranyl hexahydrate to skin areas of between 0.5 and 16 cm<sup>2</sup>) by Lopez et al. (2000) confirm the observations of earlier studies of acute exposure citing histological alterations of the kidney that increased in severity with the magnitude of exposure. Parameters describing dermal absorption coefficients for various compounds have not been reported although studies indicating changes in skin permeability with exposure to uranium (thereby favouring the entry of uranium into the body) were cited by Lopez et al. (2000).

## **7.6 Excretion and elimination**

Given the relatively low uptake of uranium from the gut it follows that most ingested *non-absorbed* uranium is excreted in faeces. Experimental evidence has shown, that once uranium enters the systemic circulation (i.e. passes through the gut wall and enters the blood) about 90% of it will be excreted through the kidneys as urine over a period of a few days. The exact proportion of uranium excreted depends upon its chemical speciation in blood. Retention of uranium in the kidney has been attributed to the creation of complexes with proteins and phospholipids in the proximal tubule (Wedeen, 1992). Faecal excretion typically accounts for less than 1% of the uranium absorbed from the gut ICRP-69, (1995a).

Clearance from the skeleton is considerably slower; half-lives of 300 and 5000 days have been estimated, based on a two-compartment model (WHO, 1998b; Kathren et al., 1989). A more in-depth discussion of the kinetics of retention following uptake into the systemic circulation is given in ICRP-69, (1995a) and/or ATSDR, (1999) to which the reader is referred.

Because of its rapid and substantial urinary excretion the concentration of uranium in urine can form the basis of assessing intake (e.g. Hooper et al., 1999; ICRP-78, 1997). Once measurements in urine have been made, biokinetic models may then be used to calculate intake. However, for inhalation, lack of information on the temporal pattern of exposure and chemical form and variation in natural excretion (e.g. Dang et al., 1992) reported urine concentrations of 0.0128 µg/l as an average for 'unexposed individuals' whereas Medley et al. (1994) observed values of 0.004 to 0.057 µg/l) can result in appreciable errors in such estimates (e.g. Stradling et al., 1998). For the specific case of DU, it may be possible to utilize differences in isotopic ratio to evaluate the upper limit of intake that corresponds to doses at the mSv level (Roth et al., 2001). Similar considerations should also apply for assessing the upper limits of DU in other tissues. These issues are discussed further in Chapters 10, 11 and 12.

For military veterans containing embedded DU, urine excretion levels of between 10 and 20 µg/l have been reported (Hooper et al. 1999). Negative finding regarding renal injury have been reported amongst such individuals (McDiarmid et al, 2000). This issue is discussed later in the following chapter. Gulf war veterans exposed to DU from inhalation, ingestion and wounds, showed average urinary excretion, 7 years post exposure, of 0.08 µg U/g creatinine, with the highest rates around 30 µg U/g (McDiarmid et al., 2000). Normal excretion of creatinine is considered to be 1.7 g/day (Jackson, 1966).

The occupational exposure decision level used for uranium workers at one facility in the United States is 0.8 µg/l of uranium in urine (FEMP, 1997). This value assumes an acute inhalation intake of moderately soluble uranium and a 60-day urine sampling frequency. For investigational purposes a value of 4 µg/l of urine is used for UK workers (information from Dr M Bailey, UK NRPB).

## **7.7 Accumulation**

In autopsies of chronically exposed individuals, uranium has been observed in the skeleton, liver and kidneys in the average ratio of 63:2.8:1 (Kathren et al., 1989). Variations in this ratio are common and are dependent on the pattern and nature of exposure (Fisenne, 1993; ATSDR, 1999). The ratios are consistent with studies performed on mine workers and members of the public (Wrenn et al., 1985) and reflect the affinity of uranium for phosphate, which is abundant in bone. A similar distribution would be expected for DU and uranium provided the patterns of intake are comparable and the delay between cessation of exposure and autopsy are similar.

In the studies performed by Pellmar et al. (1999a) rats were surgically implanted with sterilized DU and/or tantalum pellets within the gastrocnemius muscle. As early as one month after pellet implantation and at subsequent sample times (six months), brain concentrations of uranium were statistically elevated in DU-implanted rats compared to controls implanted with tantalum (e.g. less than 2 to approximately 120 ng U/g tissue after six months implantation with 20 DU 1×2 mm pellets). The authors also observed that levels of uranium were statistically elevated in the testes of exposed animals when compared to those in the control group (e.g. less than 50 to approximately 600 ng U/g tissue after 18 months implantation with 20 DU 1×2 mm pellets). Levels of uranium in both testes and brain tissues were observed to be positively correlated with exposure (number of implanted pellets). Significant amounts of uranium were excreted in urine throughout the study (e.g.  $1010 \pm 87$  ng uranium per ml urine in high-dose rats at 12-months exposure). The study suggests that in a rat model, uranium can accumulate

within the central nervous system and testicles. The accumulation of uranium in brain tissues has also been observed by Ozmen and Yurekli (1998).

No treatment-related effects (brain lesions) were identified during histopathological analysis of brains from animal studies performed by Gilman et al., (1998a, 1998b, 1998c).

## **7.8 Summary**

Uranium may enter the body through the skin, lungs or gut. Once it has entered the systemic circulation it is distributed throughout the body, where it may become absorbed onto the surface of bone, accumulate or most likely be excreted through the kidneys.

Absorption via the inhalation route depends upon the size and chemical composition of the inhaled particulates and their biological solubility. While absorption through the gut and skin largely depend upon the bioavailability of the various DU compounds to which an individual has been exposed. Typical gut absorption factors for uranium in food and water are in the order of 2% for U(VI) compounds and less for the generally more insoluble compounds of U(IV). Soluble uranium and DU compounds may be absorbed through the skin.

A number of biokinetic models exist that describe and model the biokinetics of uranium and hence DU in the body. The most recent are the ICRP models for the lung, systemic circulation and gut (all summarized in Annex 4). Whilst these models describe the distribution of uranium amongst major organs, they tend to be orientated to radiological protection issues and have not addressed more recent data relating to the distribution of uranium into testes and brain.

Current gaps in knowledge include:

- Distribution (modelled and experimentally determined) of uranium at minor concentrations and in minor organs.
- Validation of animal data to man on biodistribution into brain, liver and gonads.
- Uranium distributions at a cellular level; bio-uptake of uranium derived from DU munitions throughout all exposure pathways in comparison with typical non-munition derived uranium and DU.