THE ROYAL SOCIETY

The health hazards of depleted uranium munitions, part II

Annexe F: Groundwater transport modelling

Louise Ander and Barry Smith (BGS)

F1 Introduction

The transport of corrosion products from depleted uranium (DU) penetrators, to a defined receptor, can be considered to be a multistage process (see Figures 9 and 10 in appendix 2) in which products containing DU are:

a) dissolved in the aqueous phase

b) transported though the soil profile

c) transported through the unsaturated zone

d) transported through an aquifer until they intercept a water well or are re-introduced into the surface environment by baseflow or springs.

Sorption, retardation and dilution occur during transport to varying degrees depending upon local geological, pedological and hydrological conditions over a variety of scales. For example, hydrogeological parameters may vary almost as much over scales of metres as they do over geological units (scale 100 of metres). Furthermore, as demonstrated for example by UNEP (2001), these parameters can vary extensively over areas of military conflict (summarised variations taken from UNEP (2001) are given later in Table F7). Because of these variations it is inappropriate to use generic cases to estimate the probability of DU transport, or indeed that of any other potential pollutant of groundwater. Despite this, as indicated in the main text a number of general statements may be made from a qualitative viewpoint with regard to the likely migration of DU into groundwater reserves and thence receptors.

Whilst understanding the limitations posed when developing generic scenarios, in this annexe we consider from a quantitative viewpoint the likelihood of DU contamination of groundwater resources in three site-specific scenarios. These have been selected to highlight the variability in the likelihood of contamination, and to demonstrate the type and depth of data required to develop even preliminary quantitative site-specific assessments.

As discussed in the main text and in Annexe A, the corrosion of penetrators produces a number of secondary phases in close proximity to the penetrator/soil, penetrator/air or penetrator/water interface.

In the last case transport is dependent on the solubility (from both a thermodynamic and kinetic perspective) of corrosion products. These factors, which predominantly control migration, are discussed elsewhere in the main text of Appendix 2 and in this annexe.

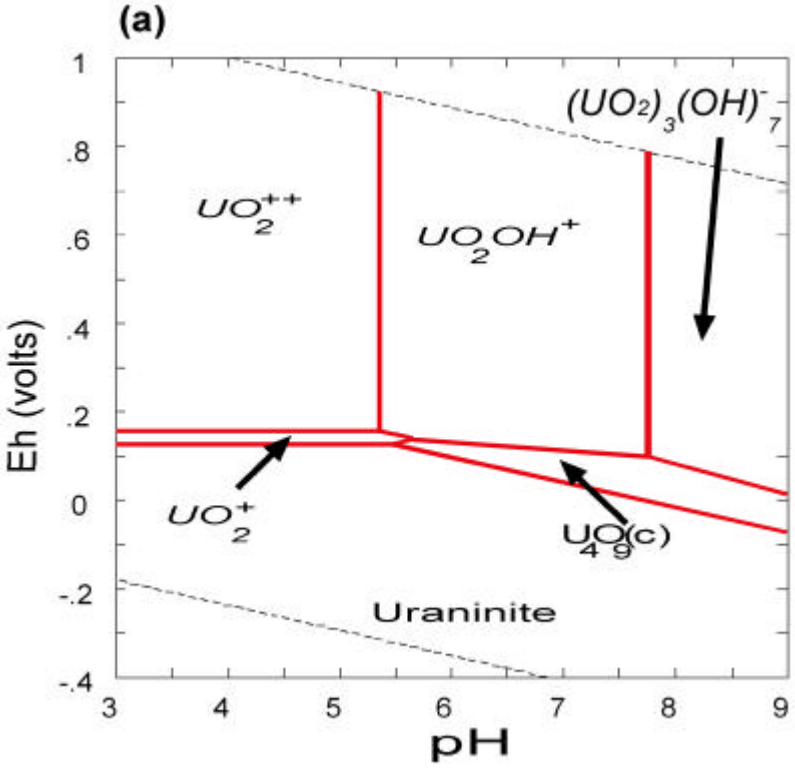
F2 Uranium solubility in natural waters

The primary control on the solubility of uranium in solution is the redox potential of the system (see chapter 4 and annexe A of appendix 2). Minerals containing U(IV) are generally very sparingly soluble (Table F1), and the mobility of any uranium introduced to a reducing environment, where U(IV) predominates, will generally be very low. However, if U(IV) is deposited in an oxidising environment, the oxidative dissolution of the mineral phases may rapidly increase the transport of uranium(VI) ions away from the source. The solubility of U(VI) is also enhanced by the presence of carbonate ions in solution, and this would be expected to increase the solubility of uranium in carbonate-buffered soil and aqueous systems.

The dominance of carbonate ions in the complexation of U(VI) species is shown in Figure F1, with a comparison of pure water and water which has been equilibriated with atmospheric carbon dioxide (pCO_2 = 0.3), resulting in dissolved inorganic carbon species. The increased solubility of uranium in the presence of carbonate ions is shown in Table F1, which compares the theoretical solubility of several common secondary uranium minerals, and shows that complexation of the uranium by carbonate greatly enhances the already more soluble U(VI) ion.

Knowledge of these likely dominant controlling factors in the solubility and transport of uranium in soils and waters can assist in the prediction of regions more likely to be vulnerable to uranium dissolution and leaching to food crops or drinking water supplies, and can assist in the prioritisation of site investigation work.

Figure F1. Eh-pH diagrams for uranium in (a) pure water and (b) in the presence of atmospheric carbon dioxide (uranium concentration = 1 µg/litre; 15 °C)



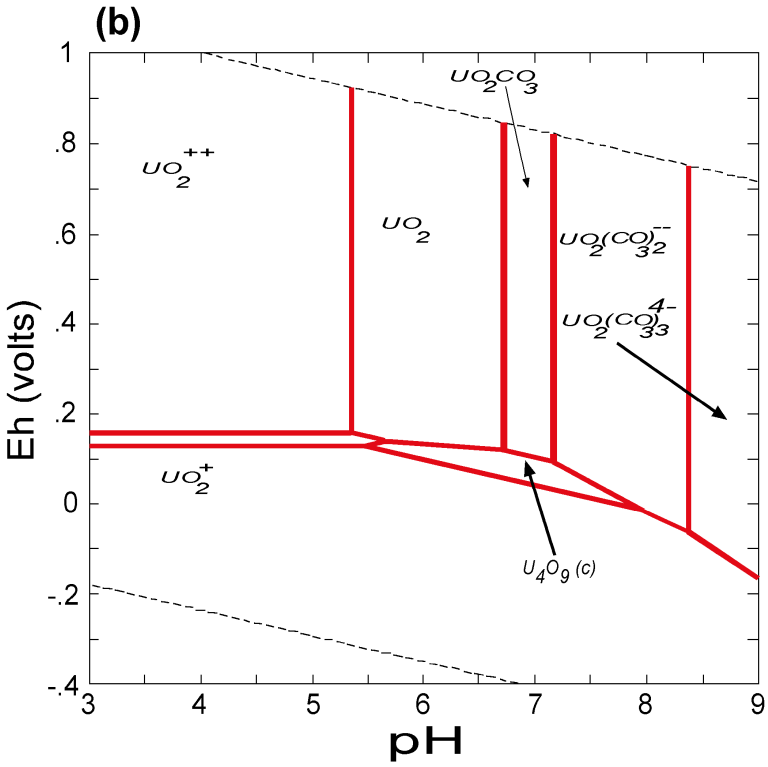


Table F1. The solubility of selected uranium minerals, at fixed temperature (15°C) and pH (7), in a range of aqueous solutions. The following simulations were run: mineral solubility in pure water; pure water in equilibrium with atmospheric CO_2; pure water in equilibrium with excess calcite; and a combination of the CO_2 and calcite models. Calculations were undertaken using PHREEQC for Windows 1.5.04 (Post 2001) using the PHREEQC-2 (version 2.4) source code (Parkhurst and Appelo 2001) and the thermodynamic data in Table F2. Each mineral of interest was allowed to reach equilibrium in solution; supersaturation of all other mineral phases was permitted for each run.

Uranium oxidation state	Mineral	Pure H_20	CO_2 equilibrium	Calcite equilibrium	Calcite and CO_2
IV	Uraninite	0.000102	0.000102	0.000102	0.000102
IV	Coffinite	0.096[1]	0.096[1]	0.140[1]	0.097[1]
IV	Ningyoite	1.70[1]	1.60[1]	18[1]	2.70[1]
VI	Soddyite	0.05	0.06	0.51[4]	0.21[5]
VI	Schoepite	1.7	2.4	25[4]	67[5]
VI	Rutherfordine	150[2]	2,700,000[3]	1200[2]	*

[1]*Actual solubility controlled by precipitation of uraninite.*

[2]*Actual solubility slightly controlled by precipitation of Schoepite.*

[3]*Actual solubility controlled by precipitation of Schoepite and U(VI) minerals.*

[4]*Actual solubility controlled by precipitation of $CaUO_4$.*

[5]*Actual solubility slightly controlled by precipitation of $CaUO_4$.*

[6]*Equilibrium not attained in calculation.*

Table F2. Thermodynamic data for dissolution of selected uranium minerals from the database llnl.dat (PHREEQC for windows 1.5.04 (Post 2001), using the source code PHREEQC-2 2.4 (Parkhurst and Appelo 2001))

Uraninite
Reaction: $UO_2 + 4H^+ = U^{4+} + 2H_2O$
log k = -4.8372
delta H = -77.8767 kJ/mol
Coffinite
Reaction: $USiO_4 + 4H^+ = SiO_2 + U^{4+} + 2H_2O$
log k = -8.0530
delta H = -1991.326 kJ/mol
Ningyoite
Reaction: $CaUP_2O_8.2H_2O + 2H^+ = Ca^{2+} + U^{4+} + 2H_2O + 2HPO_4^{2-}$
log k = -29.7931
delta H = -1016.646 kJ/mol

Soddyite Reaction: $(UO_2)2SiO_4.2H_2O + 4\ H^+ = SiO_2 + 2UO_2^{2+} + 4H_2O$ log k = 0.3920 -delta H = 0 kJ/mol
Schoepite Reaction: $UO_3.2H_2O + 2H^+ = UO_2^{2+} + 3H_2O$ log k = 4.8333 delta H = -1826.100 kJ/mol
Rutherfordine Reaction: $UO_2CO_3 + H^+ = HCO_3^- + UO_2^{2+}$ log k = -4.1064 delta H = -1689.530 kJ/mol

F3 Simulation of uranium mobility in the surface environment

To simulate the effect of different environmental conditions (pedology, geology, hydrology) on the transport of uranium from DU penetrators-contaminated soil to a groundwater receptor (here a domestic water well), simple models have been constructed. These illustrative scenarios are designed only to provide examples of the effect of different environments on uranium mobility, and hence the vulnerability of groundwater to contamination. This work does not provide site-specific information for any site impacted by DU penetrators.

ConSim (Contamination impact on groundwater – simulation by Monte Carlo method) (Environment Agency 1999) has been chosen for the purposes of this simple modelling exercise. This is a risk assessment program designed to produce probabilistic estimates of the impact of a contaminated site on groundwater; further description of the programme, can be found in Environment Agency (1999), Butler and Petts (2000), and Hawkins and Hooker (2001). In order to simplify the comparison between the different results of the simulations, the 'site' physical parameters have been kept constant where possible. The variable parameters have been chosen to represent three different scenarios, either directly or by proxy, that may be considered at the opposite ends of the uranium mobility spectrum.

F3.1 Description of conceptual models used for ConSim modelling

Two different environments have been conceptualised for the three models that represent the 'best-' and 'worst-case' scenarios summarised in Table F3. These scenarios are designed with the vulnerability of groundwater as the receptor of concern and are based on a sound understanding of the (geo)chemical controls on uranium mobility (eg Langmuir 1997), as summarised above and in the main text. It should be clear that, when the soil environment protects groundwater, the reservoir of uranium in the soil is not diminished greatly, and this may have implications for other receptors. These situations will not be dealt with in these simulations.

The best-case scenario (Uranium1) represents the best case in relation to groundwater vulnerability, ie the uranium undergoes little chemical reaction from the U(IV) solid phases derived from the oxidation of uranium metal (U(0)) and in a low permeability matrix the reaction products are not transported very rapidly away from the penetrator site. The worst case represents a much more permeable soil, which also allows oxygen ingression further into the aquifer, with inherently greater permeability allowing greater translocation of reaction products. The worst case is used in Uranium2, and further modified by Uranium3. These models are described in more detail below.

Table F3. Geochemical and Physical Environments.

	Worst case	**Best case(s)**
Geochemical parameters		
pCO_2	H	L
pH	H	L
pO_2	H	L
OC_{solid}	L	H
k_d	L	H
Physical parameters		
Flow volume	L	H or vL
Soil porewater dilution	L	H or vL

Qualitative assessment of the impact of changes in the values of the listed parameters on the mobility (H = high, L = low) of uranium in the weathering environment.

F3.2 Physical description of the contaminated 'site'

The physical geometry of the 'site' is kept constant for all the simulations, to allow the effects of the changing physicochemical weathering environment on uranium mobility to be compared more readily. The physical geometry can be summarised as follows:

source (soil) term:	10 m x 10 m x 1 m (depth)
unsaturated zone:	10 m x 10 m x 4 m (depth)
aquifer:	10 m x 10 m x 5 m (depth)
distance to receptor:	20 m in the *x* direction; lateral offset (*y*) = 0 m
hydraulic gradient:	0.001 (1 m per 1 km).

These factors are used in the parameterisation of ConSim as described below (Table F4) and are constant (unless otherwise noted) for all three scenarios. They have also been chosen to be 'single' values, rather than 'triangular', 'uniform' or 'normal' probability distributions; again this has the effect of increasing the influence of the parameters that vary between Uranium1, 2 and 3. The receptor (domestic water well) has been chosen as occurring only 20 m down flowlines

from the source; this is felt to be representative of sites observed by UNEP (2001) in former conflict areas (see Table F7 later).

Table F4. Physical parameters held constant for all 'Uranium' ConSim simulations (Environment Agency 1999)

		Uranium1-3
Soil zone		
Width	m	SINGLE(10)
Length	m	SINGLE(10)
Thickness	m	SINGLE(1)
Width perpendicular to flow	m	SINGLE(10)
Unsaturated zone		
Infiltration rate	mm/a	SINGLE(200)
Thickness	m	SINGLE(4)[2]
Vertical dispersivity[1]	m	SINGLE(0.04)[2]
Aquifer zone		
Hydraulic gradient	-	SINGLE(0.001)
Vertical dispersivity[1]	m	SINGLE(0.2)
Longitudinal dispersivity[1]	m	SINGLE(2)
Transverse dispersivity[1]	m	SINGLE(1)
Distance to receptor (*x*)	m	SINGLE(20)
Distance to receptor (*y*)	m	SINGLE(0)
Simulations		1000

[1]*Dispersivity values have been calculated as scale-dependent variables of site geometry, as discussed in Appelo and Postma (1994).*

[2]*Uranium3 uses a UNIFORM(0.5,1) thickness and UNIFORM(0.005,0.01) dispersivity.*

F3.3 Descriptions of illustrative scenarios

F3.3.1 Uranium1 (Best case) parameterisation

The variable physical parameters for this simulation have been chosen using representative published values (Table F5). The conceptualised environment is that of a clay-rich soil overlying mudrock aquifer, including a weathered zone as the unsaturated zone and presuming that there is no secondary fracture flow . Thus, values appropriate to these environments have been used, resulting in low permeability and conductivity values (which will have most effect on the velocity of weathering product translocation).

In establishing appropriate values to use for the chemical input of Uranium1, the following considerations have been made. This environment has a high clay content, low redox potential

(low oxygen ingression through the upper part of the soil) and moderately low pH. Because ConSim is not designed to deal with geochemical input values, such as pH, the partition coefficient (K_d) has to be used as a proxy for all the above features; an example of this can be seen from the variation in K_d with pH shown in Figure F2. In the environment described here, the K_d value would be expected to be very high (ie the uranium is found associated with the solid phases of the system not the aqueous phases). The issues arising from the use of K_d values will not be discussed here, but have been summarised by publications such as Reardon (1981), Bethke and Brady (2000), and USEPA (1999). The values for K_d have been taken from the experimental results of Zachara et al (1996) in USEPA (1999); these experiments are cited as having been undertaken using clay minerals in the absence of dissolved carbonate, which is analogous to the environment being modelled here. The summary of their results is shown in Figure F3, from which the distribution of K_d values used in Table F6 have been derived.

Figure F2. Variation in K_d with soil pH (reproduced from USEPA (1999)).

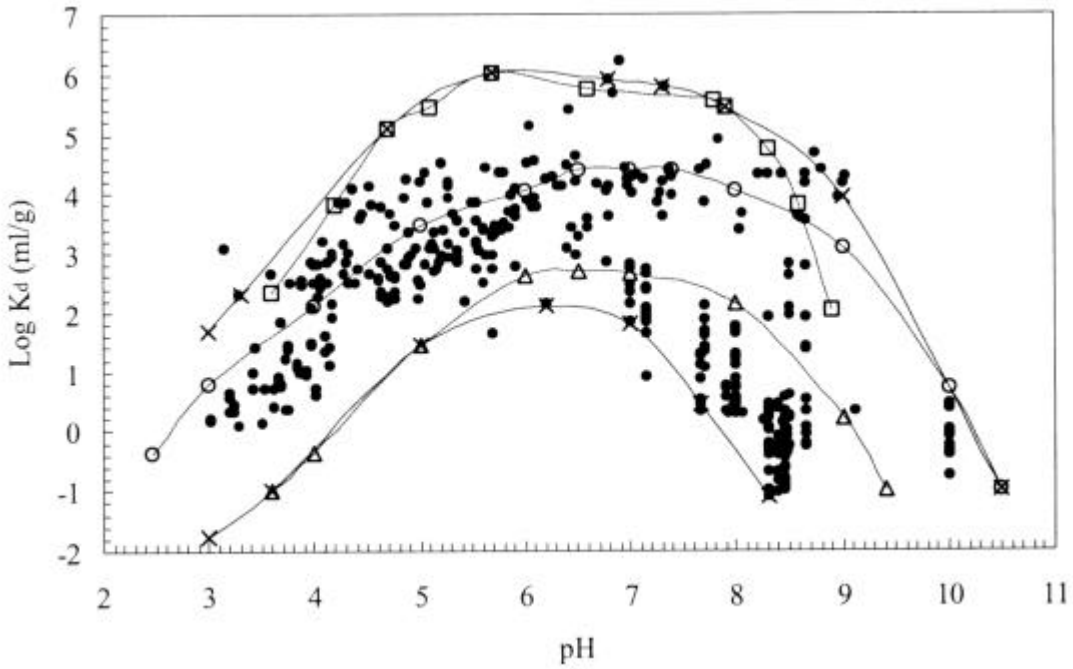
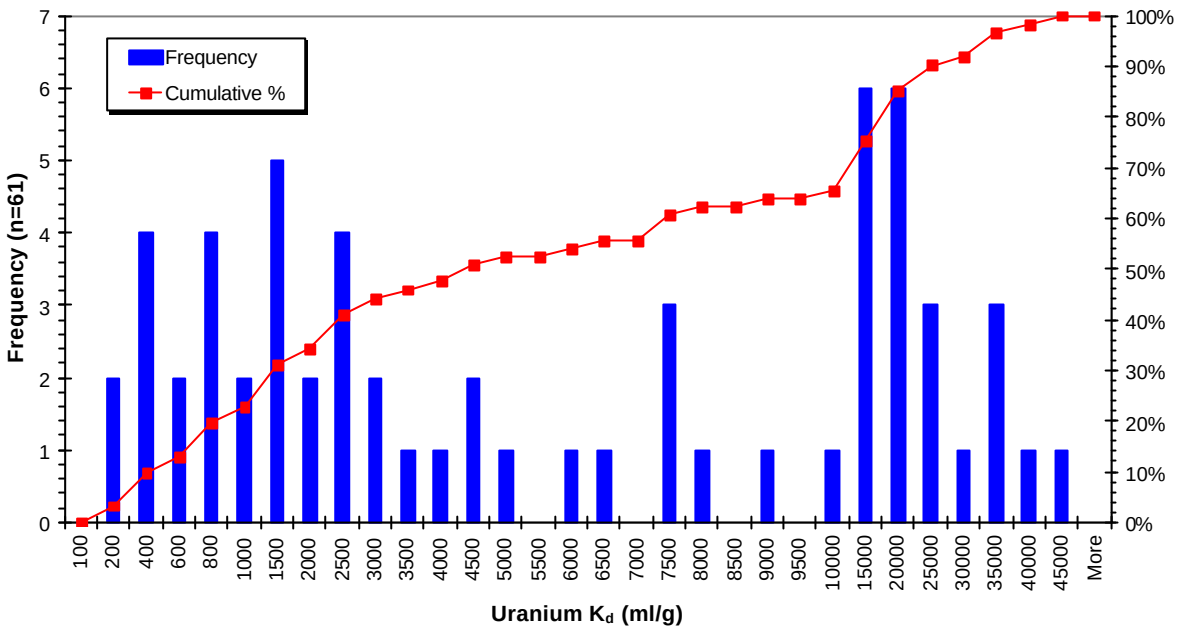


Figure F3. Variation in K_d for clay (argillaceous) soils (derived from USEPA 1999)).



In assessing the maximum likely concentration of uranium in these waters (Table F6), the results of simple equilibrium modelling in PHREEQC-2 have been used (Table F1). The environment simulated here is closest to that of uraninite stability (ie U(IV) mineral). The chosen value for uranium solubility in equilibrium with a hypothetical CO_2-free environment has been used (Table F1).

ConSim also requires an estimate of the soil concentration as the only measure of the source term that can be input. Again, it has been presumed that the weathering of the penetrators (subsurface) is less than that which would take place in a more oxidising environment (Uranium2), so the values from Tables 3 and 4 in Annexe A have been used from the lower weathering rates, and therefore lower concentrations occur in the soil, in a form that is available for leaching.

The background concentration of uranium in groundwater is chosen as a constant, very low value, of 0.00002 mg/l in all models (Table F6); however, it should be recognised that natural variability in uranium concentrations can be considerable (WHO 2001).

The model being demonstrated here is an environment with a low rate of uranium weathering, coupled with low rates of groundwater flow, resulting in very slow transport timescales to the receptor being expected.

F3.3.2 Uranium2 (worst case) parameterisation

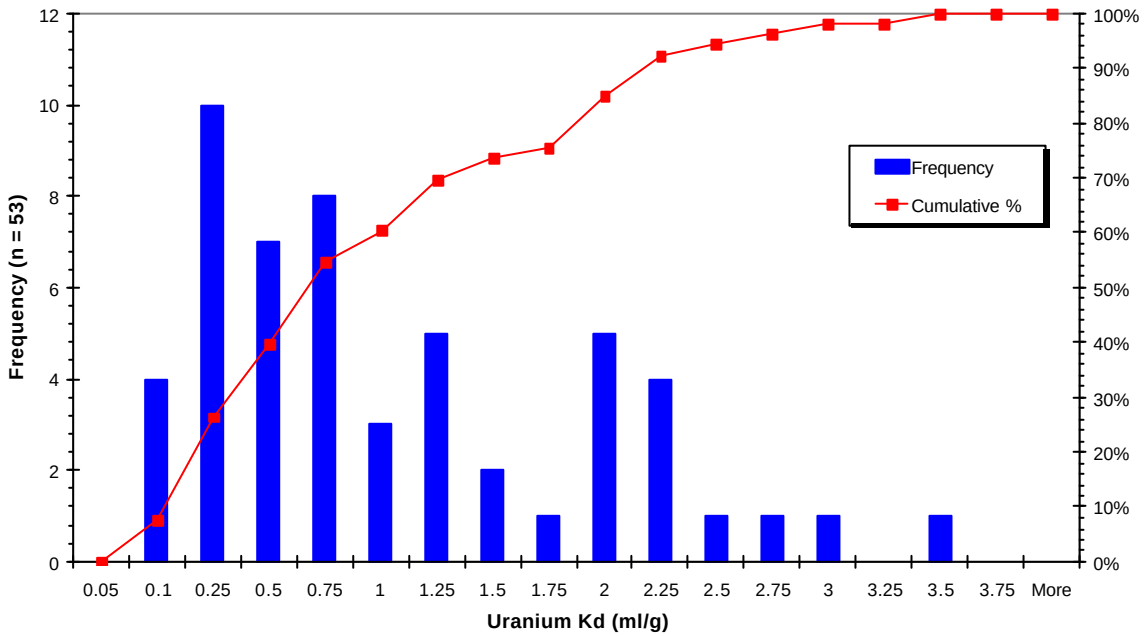
The scenario-specific physical parameters (Table F5) have been derived from published values for sand/sandy loam soils, with an unsaturated zone comprised of weathered sandstone, overlying

the sandstone aquifer. It has been presumed that the sandstone has a high primary permeability and that preferential fracture flow (which would increase flow velocity) does not occur.

The soil and underlying strata are sufficiently permeable to allow rapid recharge, and the waters are all oxidising, suggesting that the uranium will occur as U(VI), with solubility controlled by schoepite (see above). Additionally, it is assumed that there is a high concentration of CO_2 in solution, and sufficient calcite in the geological materials to maintain an excess of bicarbonate ions in solution to complex with uranium, further increasing the solubility of uranium. The implication of these comments is that the pH must be alkaline. All of these conditions suggest that the K_d used should be very low. The source of K_d values used (Figure F4; Table F6) is the results of Kaplan and Serne (1995) and Kaplan et al (1996,1998) in USEPA (1999), where their work is reported to have been undertaken with arenaceous materials, reflecting a similar environment to that chosen here.

The maximum solubility of uranium in this model has been derived from the PHREEQC modelling described above (Table F6), for an oxidising, high pCO_2 environment. The soil (source) concentration term has been derived presuming the most rapid rates of weathering of DU penetrators calculated in Tables 3 and 4 in Annexe A, which is also designed to reflect the geochemical environment being simulated.

Figure F4. Variation in K_d for sandy (arenaceous) soils (derived from USEPA (1999))



The environment being parameterised here is designed to represent a carbonate-rich oxidising aquifer where uranium solubility is increased by the presence of U(VI) carbonate aqueous species, and where flow rates are sufficient to remove all the soluble reaction products. These result in a more rapid transfer of reaction products (aqueous uranium) to the receptor than is the case in Uranium1.

F3.3.3 Uranium3 parameterisation

This simulation is identical to Uranium2 with the exception of the values used for the unsaturated zone thickness and partition coefficients (K_d values). The unsaturated zone has been made variably thinner (Table F5) to represent an even more vulnerable situation (in relation to groundwater) than found in Uranium2. The values of K_d have been reduced to more realistically represent the situation that would be found in a grossly contaminated scenario, where all the potentially available surface sorption sites in the soil matrix are occupied and no sorption can occur; thus K_d is zero in the soil. In the unsaturated zone, the K_d value has also been reduced from that used in Uranium2, to account for a high loading of uranium of potentially available surface sites, again reducing sorption potential. The K_d value of the aquifer material has been left unchanged from the Uranium2 scenario. This may represent a more realistic situation in a carbonate-rich oxidising soil-aquifer system which has been contaminated for a duration of several years, than does the consistent K_d used in Uranium1 and Uranium2.

The model being described here is that of a high weathering environment, which has been contaminated for a duration of several years, coupled with more rapid transport of reaction products to the groundwater system.

Table F5. Variable physical parameters for ConSim (Environment Agency 1999)

		Distribution	**Uranium1**	**Uranium2**	**Uranium3**
Soil zone					
Water-filled porosity[1]	fraction	UNIFORM	0.05,0.15	0.05,0.15	0.05,0.15
Air-filled porosity[1]	fraction	UNIFORM	0.15,0.25	0.15,0.25	0.15,0.25
Bulk density[2]	g/cm^3	NORMAL	1.3,0.4	1.4,0.2	1.4,0.2
Unsaturated zone					
Bulk density[2]	g/cm^3	TRIANG	1.7,2.03,2.35	1.6,2.68	1.6,2.68
Effective porosity[3]	fraction	UNIFORM	3,5	0.2,0.35	0.2,0.35
Hydraulic conductivity[2]	m/s	TRIANG	1e^{-13},1e^{-10}, 1e^{-9}	3e^{-10},1e^{-7}, 6e^{-6}	3e-^{10},1e^{-7}, 6e^{-6}
Aquifer zone					
Effective porosity[3]	fraction	UNIFORM	0.005,0.05	0.005,0.01	0.005,0.01
Bulk density[2]	g/cm^3	UNIFORM	1.54,3.17	1.6,2.68	1.6,2.68
Hydraulic conductivity[2]	m/s	TRIANG	1e^{-13},1e^{-10}, 1e^{-9}	3e^{-10},1e^{-7}, 6e^{-6}	3e-^{10},1e^{-7}, 6e^{-6}

[1]*Taken from ConSim demonstration files as reasonable.*

[2]*ConSim tabulation of lithology specific values.*

[3]*Domenico and Schwartz (1990).*

Table F6. Chemical input parameters for ConSim (Environment Agency 1999)

		Distribution	**Uranium1**	**Uranium2**	**Uranium3**
K_d uranium[1]					
soil	ml/g	TRIANG	500,400,20000	0.05,0.5,3.5	[SINGLE(0)]
unsaturated zone	ml/g	TRIANG	500,400,20000	0.05,0.5,3.5	0,0.005,0.05
saturated zone	ml/g	TRIANG	500,400,20000	0.05,0.5,3.5	0.05,0.5,3.5
Soil concentration	mg/kg	TRIANG	0.05,0.2,0.5	0.2,0.4,1.3	0.2,0.4,1.3
Solubility	mg/l	SINGLE	0.0001	2.4	2.4
Background aqueous concentration	mg/l	SINGLE	0.00002	0.00002	0.00002

[1]*USEPA (1999); for further description see text above.*

F3.4 Results

The results below demonstrate the significant difference in translocation of uranium weathering products to groundwater receptors, depending on the pedological, geological and hydrological environment based on the simple models described above. These results reinforce the comments made above, regarding the requirement to undertake site-specific studies where receptors occur close to areas of conflict in which DU penetrators have been used.

F3.4.1 Uranium1

The most likely transit time for migration to reach a drinking water well sited 20 m from a DU strike site is around 30 million years (range 1 million to 100 million years) for the best-case scenario (Uranium1). The most likely concentration of DU at the point of water abstraction after an elapsed time of 1 million years was << 0.1 µg/litre (note limit of model predictor = 1 million years). After a period of 1 million years the model predicted < 0.1 µg/litre DU in pore fluids at the base of the unsaturated zone.

F3.4.2 Uranium2

The most likely transit time for migration to reach a drinking water well sited 20 m from a DU strike site is about 110 years (range 25 to 350 years) for the worst-case scenario (Uranium2). The most likely concentration of DU at the point of water abstraction after an elapsed time of 110 years was 20 µg/litre (range < 0.1 to 400 µg/litre). Peak concentrations of DU in pore fluids at the base of the unsaturated zone reached 2.4 mg/litre after a period of 20 years and gradually decreased to 0.5 mg/l after 100 years.

F3.4.3 Uranium 3

The most likely transit time for migration to reach a drinking water well sited 20 m from a DU strike site is about 60 years (range 10 to 350 years) for the worst-case scenario (Uranium2). The most likely concentration of DU at the point of water abstraction after an elapsed time of 60

years was 30 µg/litre (range < 0.1 to 500 µg/litre). Peak concentrations of DU in pore fluids at the base of the unsaturated zone reached 2.4 mg/l after a period of 2 years.

Reduction in the depth of the unsaturated zone and the soil K_d used for the Uranium3 scenario also represent a situation in which DU projectiles penetrate deep into the soil, thereby bypassing surficial soil layers, which being richer in organic carbon often exhibit higher K_d values for uranium. Similarly, in such a situation the distance through which dissolved DU has to migrate through the unsaturated zone will be decreased. In such circumstances the results from the scenario for Uranium3 predict the rapid transport of DU to the base of the saturated zone with little retardation or dilution.

The results for Uranium2 and Uranium3 suggest that in an environment similar to that modelled here, a domestic water well may be at risk of exceeding the recommended WHO maximum in drinking water of 0.002 mg/l uranium. In the conservative clay-rich, hi-K_d environment modelled in Uranium1, little uranium is predicted to be mobilised, and therefore is not predicted to reach the receptor in measurable quantities even after a million years have elapsed. However, it should be noted that this is only a hypothetical case study, and does not represent physical situations, such as fracture networks in a mudrock aquifer, where fluid flow rates would be greater. In all situations encountered in former conflict areas, site-specific investigations are prerequisites to establishing likely transport of uranium to aqueous receptors, such as local groundwater resources.

Figure F5(a). Uranium1: the retarded time for travel is very high in this simulation, showing that were these conditions (or those close to them) to prevail in an affected area, the defined receptor is unlikely to be affected (reverse cumulative probability plot, 1000 trials).

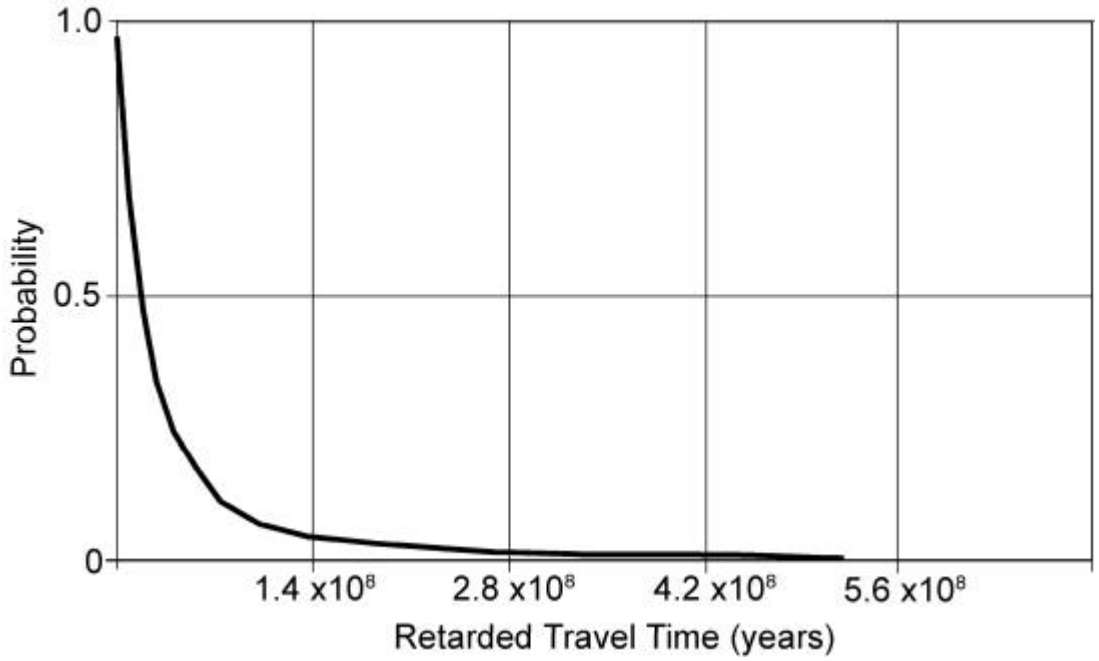


Figure F5(b). Uranium2 model retarded travel time to the receptor (reverse cumulative probability plot, 1000 trials). Note the extreme difference in the retarded travel time in comparison with Figure F4(a).

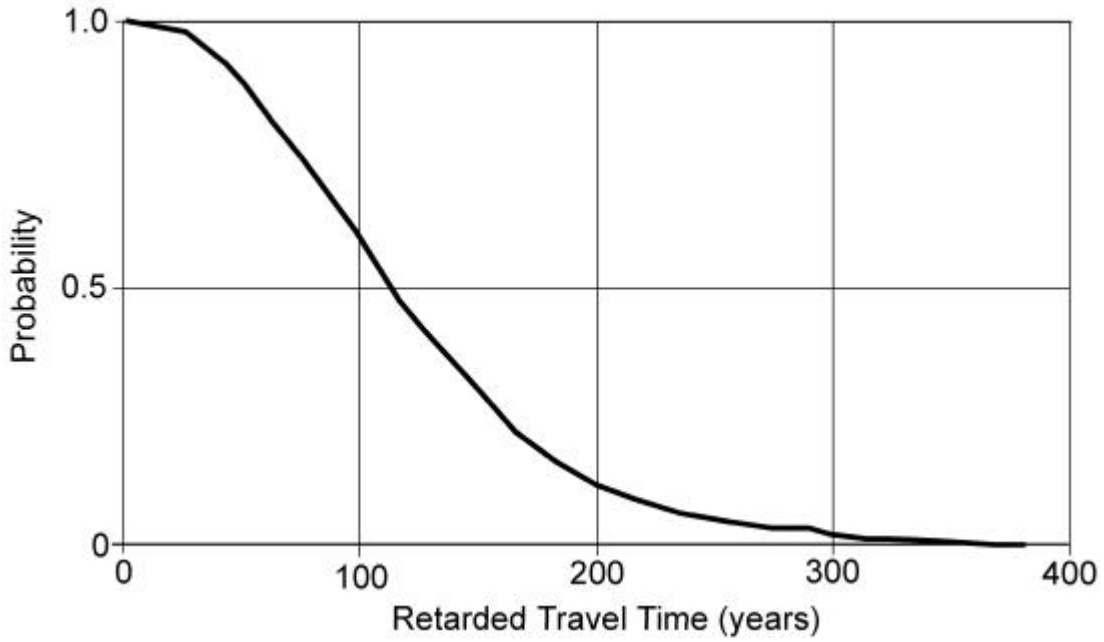


Figure F5(c). Uranium3 model retarded travel time to the receptor (reverse cumulative probability plot, 1000 trials).

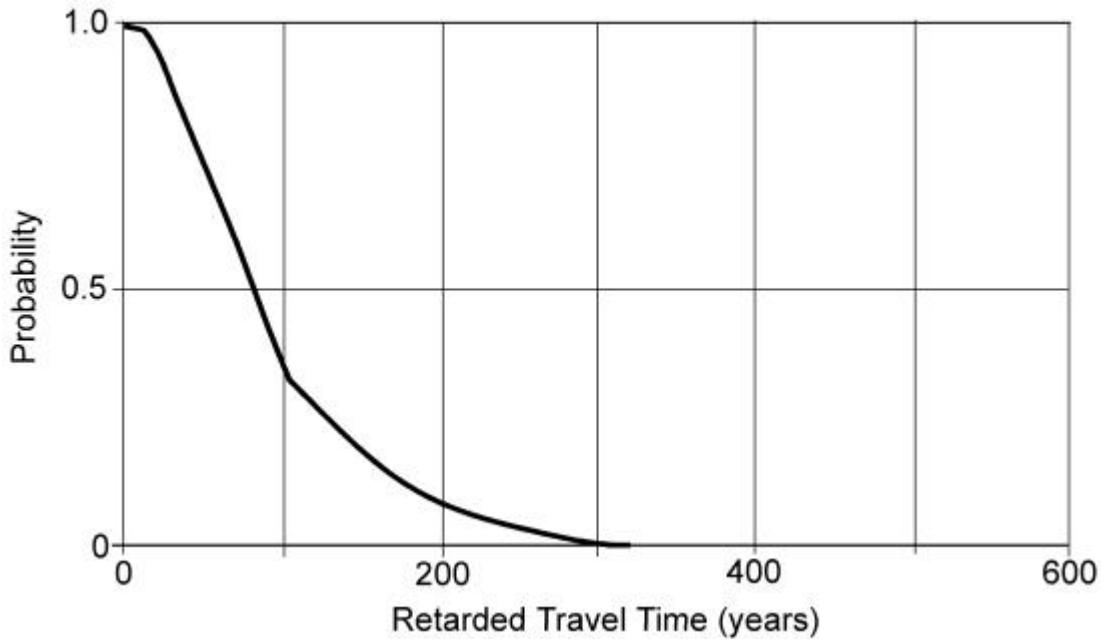


Figure F5(d). Uranium2. This figure shows that in this model the concentration of uranium in solution is likely to exceed the WHO recommended limit (0.002 mg/l) in a decadal timeframe, suggesting that continued monitoring of the water well would be required over a long timeframe to ensure that human receptors were not affected.

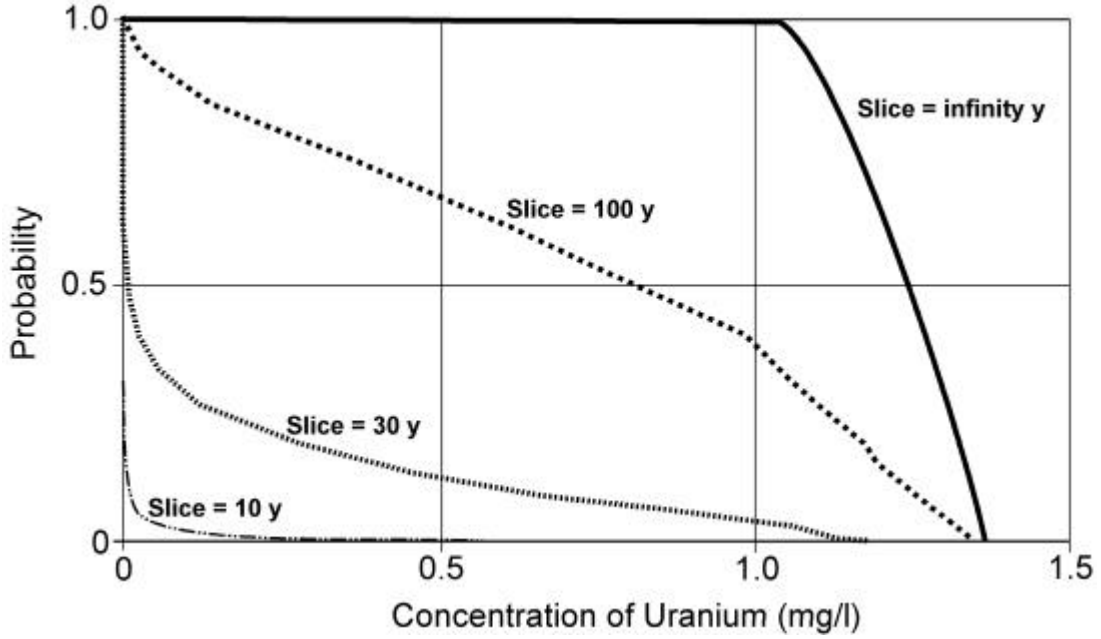


Table F7. Summarised estimates of hydraulic properties for strike sites in Kosovo, from UNEP (2001)

Planeje Village
< 10 cm soil cover
Geology
Groundwater level 2 m below ground level
Karstic?
Supersaturated with respect to calcite
Hydraulic conductivity limestones = 10^{-5} to 10^{-9}
Porosity 0.1
Head 0.5 m
Receptors 50 m
Estimated K_d aquifer 1000
Vranoc
Glacial ridge 30 m above surrounding landscape
Groundwater level 2 m below ground level at foot of esker
Estimated fine sand K_d 4-200
Humus layer 0.5 m
Hydraulic conductivity silty sand 10^{-3} to 10^{-7}
Porosity 0.3
Head 1m
Receptors 250 m

Irziniq Karst Receptors 500 m Groundwater level 5 m below ground level Soil 4 m deep Under saturated with respect to calcite Reducing Eh conditions? Hydraulic conductivity = 10^{-2} to 10^{-6} Porosity 0.4 Head 0.1
Bellobrade Groundwater level 2 m below ground level Receptor 1000 m Undersaturated with respect to calcite Geology unknown Fine-grained sand assumed Hydraulic conductivity 10^{-2} to 10^{-5} m/s Porosity 0.4 Head 0.5 m
Bandera GWL 11 bgl Undersaturated with respect to calcite Reducing Eh conditions? Overall receptor 500 +/- 300 m

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