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LLWR Lifetime Project

Sorption Parameters for the LLWR Geosphere
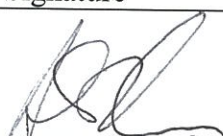
A report prepared by Nexia Solutions for and on behalf of the Low Level Waste Repository Site Licence Company

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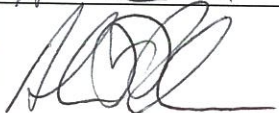## *LLWR Lifetime Project:*

Sorption parameters for the LLWR geosphere

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LLWR Lifetime Project: Sorption parameters for the LLWR geosphere

Nexia Solutions (08) 9451
Issue 1

Matt Randall, September, 2008

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EXECUTIVE SUMMARY

The Environment Agency (the Agency) is the UK regulatory authority responsible for authorising the disposal of radioactive waste at the Low Level Waste Repository (LLWR) in West Cumbria. The Agency issued a new Authorisation for the LLWR on 1 May 2006, which includes a list of improvement and additional information requirements that must be fulfilled by the operator (Schedule 9 of the authorisation). These requirements are being addressed within the LLWR Lifetime Project. The LLWR Site Licence Company (LLWR SLC) is responsible for the management and ownership of the project, as well as its technical direction.

An interim radiological assessment for the groundwater pathway has been undertaken within the LLWR Lifetime Project, building on the assessments undertaken for the 2002 safety cases. A key element of the groundwater pathway assessments is an understanding and simulation of the transport of radionuclides through the geosphere. Sorption processes play an important role in controlling this travel time and so the specification of appropriate parameters representing these sorption processes is a key task.

This report provides a description of the derivation of appropriate sorption parameters, or K_ds, for use within the groundwater pathway calculations. It builds on previous work undertaken as part of the 2002 Post Closure Safety Case (PCSC), taking into account recent experimental work and a review of the existing underpinning experimental data.

Several changes have been made to the sorption data, compared to the values used in the 2002 PCSC. These changes have been mainly due to a closer examination of the experimental protocols used in site specific sorption experiments and tighter criteria for the relevant groundwater conditions. In particular, only sorption measurements carried out at pHs relevant to the prevailing groundwater conditions have been considered in the derivation of K_ds.

The sorption behaviour of uranium has been a particular focus of work undertaken post 2002. This has included experimental work to further elucidate the variability and controlling mechanisms associated with the sorption of this radionuclide. Additionally, a simple surface complexation model has been successfully applied to the experimental data. This model is able to reproduce the sorption of uranium at a range of geochemical conditions and for a number of sorption substrates. The success of this model means that estimates of variability of uranium sorption in the LLWR geosphere can be made based on a good understanding of the underpinning sorption mechanisms.

An updated set of K_ds as used in the current radiological assessment is presented within this report. These K_ds are underpinned by site specific measurements. Where possible, an indication of the variability of these parameters is also presented within the report. This can inform future evaluations of the influence of uncertainties in K_ds on radiological assessment results for the groundwater pathway.

PREFACE

This report has been produced by Nexia Solutions Ltd in collaboration with and on behalf of the Low Level Waste Repository Site Licence Company in line with the agreed LLWR Lifetime Project strategy and philosophy and the inclusion of technical data concerning the LLWR operations and site evolution strategy.

VERIFICATION STATEMENT

This document has been verified and is fit for purpose. An auditable record has been made of the verification process. The scope of the verification was to confirm that : -

• The document meets the requirements as defined in the task specification/scope statement.
• The constraints are valid.
• The assumptions are reasonable.
• The document demonstrates that the project is using the latest company approved data.
• The document is internally self consistent.

HISTORY SHEET

Issue Number	Date	Comments
Draft 1	27/03/08	For checking
Issue 0.1	31/03/08	Verified and approved by Nexia Solutions
		Issued to LLW Repository Ltd. for comment
Issue 0.2	21/07/08	Updated to address comments on Issue 0.1. Issued to LLW Repository Ltd.
Issue 1	17/09/08	Approved by the LLWR and raised to Issue 1. No other amendments made.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

1.   Introduction

1.1.   Background

The Environment Agency (the Agency) is the UK regulatory authority responsible for authorising the disposal of radioactive waste at the Low Level Waste Repository (LLWR) in West Cumbria. The Agency issued a new Authorisation for the LLWR on 1 May 2006, which includes a list of improvement and additional information requirements that must be fulfilled by the operator (Schedule 9 of the authorisation). These requirements are being addressed within the LLWR Lifetime Project. The LLWR Site Licence Company (LLWR SLC) is responsible for the management and ownership of the project, as well as its technical direction.

The authorisation issued in 2006 followed a review of the previous authorisations for the site and was informed primarily by a review of the 2002 Operational Environmental Safety Case (OESC) and Post-Closure Safety Case (PCSC) submitted by British Nuclear Fuels Ltd (BNFL) (BNFL, 2002a,b). As part of the LLWR Lifetime Project, an interim radiological assessment for the groundwater pathway has been undertaken to support the submission against Requirement 2 of Schedule 9 of the current authorisation (Paksy and Henderson, 2008). This builds on the assessments undertaken for the 2002 safety cases. This report provides a description of the derivation of appropriate sorption parameters, or K_ds, used within the groundwater pathway calculations. It builds on previous work undertaken as part of the 2002 PCSC, taking into account recent experimental work (Dutton and Trivedi, 2008) and a review of the existing underpinning experimental data.

As described in Paksy and Henderson (2008), the radionuclides included within the radiological assessment for the groundwater pathway are:

Am-241, Am-242m, Am-243, C-14, Cl-36, Cm-243, Cm-244, Cm-245, Cm-246, Cm-248, Co-60, Cs-135, Cs-137, H-3, I-125, I-129, I-131, Nb-94, Nb-95, Ni-63, Np-237, Pa-231, Pb-210, Pu-236, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Ra-226, Sr-90, Tc-99, Th-230, Th-232, U-233, U-234, U-235, U-238, Zr-93

Sorption data are therefore presented for all of the radioelements listed above.

1.2.   Report structure

This report is structured as below:

- Section 1 is this introduction.

- Section 2 describes the representation of sorption in assessment models and presents the data used in the 2002 PCSC.

- Section 3 presents an overview of the LLWR geosphere characteristics, including the geology, mineralogy and groundwater chemistry.

- Section 4 provides a review of sorption for radionuclides considered in the radiological assessment undertaken to support the submission against Requirement 2 of Schedule 9 of the current authorisation (Paksy and Henderson, 2008) and the specification of appropriate sorption parameters for use in the radiological assessment.

2. Sorption in the geosphere

2.1. Overview of sorption processes

Sorption processes play an important role in controlling the time taken for dissolved species to travel through the geosphere. Sorption, which can be defined as the accumulation of matter at the interface between the solid surface and the aqueous phase, includes ion-exchange, where the interaction is controlled primarily by electrostatic attraction onto fixed charged sites, and surface complexation onto variable charge sites. When combined with advective and diffusive transport, sorption processes result in a net retardation of a chemical substance relative to a conservative non-sorbing tracer in the aqueous phase.

At the molecular scale, sorption processes can be very complex and by necessity the representation of sorption processes within models involves the use of simplifications and assumptions. The most commonly used method to represent sorption within an assessment level model is the specification of a distribution coefficient, or K_d, for each element of interest.

K_d is a term that is based on thermodynamic principles such as instantaneous sorption, reversibility and a linear relationship between the amount of solute on the solid phase and the concentration of the solute in solution. It is not always possible to demonstrate these criteria, particularly in natural systems, and so the sorption coefficients derived from complex natural or experimental systems are more correctly termed a distribution coefficient, or R_d, defined as:

$$R_d = \frac{Radionuclide\ concentration\ in\ the\ solid\ phase}{Radionuclide\ concentration\ in\ the\ liquid\ phase}$$

In this report it is assumed that measured R_d values provide a good estimate of the linear equilibrium K_ds that are assumed in the radiological assessment calculations.

2.2. Sorption data used in the 2002 PCSC

The sorption data used in the 2002 PCSC were derived from a consideration of site specific sorption experiments that were deemed appropriate to the conditions expected to be encountered within the geosphere and is described in full in Randall (2002). In general terms, sorption parameters were derived by calculating the geometric mean of these site specific data and were presented separately for coarse, silt and clay sediments. The three classes are defined below.

- Coarse sediment: This must contain more than 65% of particles with particle size greater than 0.06 mm (i.e. sand, gravel and cobble). For guidance but not as part of the definition, such sediments may contain up to 5% silt, < 1% organic matter and < 1% clay.

- Silty sediment: This must contain less than 65% sand and larger particles, and < 35% clay. For guidance, it should contain more than 5% silt and between 1-10% organic matter. This definition allows for a wide range of textures.

- Clay-rich sediment: This must contain > 35% clay. It may contain up to 50% organic matter and varying amounts of silt and sand.

The parameters derived within Randall (2002) are presented in Table 1.

Table 1: Summary of recommended K_d values ($m^3 kg^{-1}$).

	Coarse			Silt			Clay		
	Value	Max	Min	Value	Max	Min	Value	Max	Min
Cs	0.30	3.2	0.07	1.2	5.1	0.06	7.5	-	-
Ca	0	-	-	0	-	-	0	-	-
Sr	0.012	0.023	0.0078	0.021	0.032	0.008	0.029	0.11	0.06
Ra	0.086	0.097	0.077	0.057	-	-	0.057	-	-
Ni	0.056	0.24	0.02	0.056	-	-	0.056	-	-
Zr	0.55	-	-	0.55	-	-	24	-	-
Nb	0.55	-	-	0.55	-	-	24	-	-
Mo	1×10^{-3}	-	-	1×10^{-3}	-	-	1×10^{-3}	-	-
Tc	1×10^{-4}	5×10^{-4}	0	1×10^{-4}	-	-	1×10^{-4}	-	-
C	0	-	-	0	-	-	0	-	-
Pb	0.056	-	-	0.056	-	-	0.056	-	-
Cl	0	-	-	0	-	-	0	-	-
I	0.0011	0.002	0.0006	0.0011	-	-	0.0011	-	-
Ac	0.12	-	-	0.12	-	-	0.12	-	-
Th	0.55	0.88	0.41	0.55	-	-	24	-	-
Pa	0.11	-	-	0.11	-	-	0.11	-	-
U	0.16	19	0.0031	0.027	-	-	0.018	0.098	0.003
Np	0.024	0.04	0.011	0.032	-	-	0.032	-	-
Pu	0.11	9	0.03	6.6	-	-	8.1	6.8	10
Am	0.12	0.17	0.08	0.12	-	-	0.12	-	-
Cm	0.12	-	-	0.12	-	-	0.12	-	-

For use in the 2002 PCSC geosphere model, K_d data for the "coarse" sediment class were considered the most appropriate for representing sorption within the drift material.

Additionally, parameters representing sorption onto the sandstone bedrock were taken from Quantisci (1999). These values are reproduced in Table 2, along with the "coarse" sediment values adopted in the 2002 PCSC for the drift material. For certain elements, such as cobalt, data for chemically analogous elements were used.

Table 2: K$_d$s (m^3 kg^{-1}) used in the 2002 PCSC.

Species	Drift	Sandstone
Ac	1.2E-01	1.0E-01
Am	1.2E-01	1.0E-01
C	0.0E+00	1.0E-03
Cl	0.0E+00	0.0E+00
Cm	1.2E-01	1.0E-01
Co	5.6E-02	1.0E-02
Cs	3.0E-01	3.0E-01
H	0.0E+00	0.0E+00
I	1.1E-03	0.0E+00
Mo	1.0E-03	1.0E-04
Nb	5.5E-01	5.0E-01
Ni	5.6E-02	1.0E-02
Np	2.4E-02	5.0E-03
Pa	1.1E-01	1.0E-01
Pb	5.6E-02	5.0E-02
Pu	1.1E-01	1.0E-01
Ra	8.6E-02	3.0E-01
Sr	1.2E-02	3.0E-01
Tc	1.0E-04	1.0E-04
Th	5.5E-01	1.0E+00
U	1.6E-01	1.0E-02
Zr	5.5E-01	5.0E-01

3.   Characteristics of the geosphere

3.1.   Introduction

The sorption of radionuclides in the geosphere is controlled, to a large extent, by the characteristics of the geology. This includes the nature of mineral surfaces encountered by the radionuclides within the geosphere and chemical conditions within the groundwater. The following sections therefore provide a summary of the geology, the mineralogy of the sediments and the groundwater conditions in the vicinity of the LLWR.

3.2.   Geology

The LLWR has a complex geological history, with up to 50 m of variable drift deposits (unconsolidated material), overlying the solid bedrock. A brief description of the geological sequence from the bedrock to the ground surface is given below.

3.2.1. Sandstone Bedrock

The Ormskirk Sandstone Formation of the Sherwood Sandstone Group forms the uppermost unit of bedrock and is predominantly aeolian in the West Cumbria area (Akhurst *et al.*, 1997). The formation has a relatively homogeneous lithological composition of weakly to firmly cemented fine to medium-grained sandstone. There are subtle variations in composition, with variations in cement strength and lithological composition recognised vertically from analysis of cores recovered during on-site drilling. The formation's aeolian facies is characterised by small, centimetre-scale, dune-bedded, cross-bedding up to metre-scale sets (BNFL, 2002c).

Large-scale faulting with a similar orientation to the regional structural orientation may exist beneath the LLWR site, but a lack of information from the deeper sub-surface means this cannot be proved or disproved.

Locally, there may also be smaller associated zones of structural disturbance. Frequent, small-scale structural discontinuities including faults, fractures and joints have been recognised at outcrop at the nearby Sellafield site. These fractures range from open to partially or completely in-filled or cemented. Their spacing varies from centimetres to metres (BNFL, 2002c).

The subcrop of the Ormskirk Sandstone Formation has its greatest elevation under the north-eastern part of the consented area with some of the superficial deposits on-lapping onto this high. At the high, the upper surface of the sandstone subcrop is 15 m below ground level (bgl). There is a gradual dip in this surface to the south-west so that it is approximately 30 m bgl at the coast. There are no sandstone exposures at the surface in the immediate vicinity of the site, although sandstone bedrock is exposed further away from the LLWR on the beach at Seascale, in stream sections inland towards Gosforth and in the River Calder at Sellafield (Hunter *et al.*, 2008).

The sandstones of the Triassic Sherwood Sandstone Group, which underlie the site and form the bedrock to all of the coastal plain of the LLWR region are several hundred metres thick (Hunter *et al*., 2008).

3.2.2. Drift Deposits

Quaternary-aged superficial deposits overlying the bedrock consist of a variable thickness of heterogeneous glacial and post-glacial sediments. The thickness of these deposits varies between 10 m at the north-west end of the trenches, 35 m between the site and the coast and a maximum of 50 m at the south-western end of the site. These deposits are mainly of glacial origin with a thin 'veneer' of post-glacial sands and peats. The latter are present within the site and between the site and the coast, and are represented by a relatively flat (compared with dunes down the coast towards the Ravenglass estuary), dune-type environment. The glacial deposits are heterogeneous in terms of texture and structure over a range of scales with deposits ranging from compacted clays to coarse sands and gravels (see Section 7.2 of BNFL, 2002c).

The following summary description of superficial deposits in the LLWR area is based mostly on the recent reinterpretations of Hunter *et al.* (2008) and Smith (2008), with some additional information from BNFL (2002c,d). The re-interpretation has redefined superficial deposits into seven units (termed Lithofacies Packages (LP) 1–7) (Hunter *et al.,* 2008; Smith, 2008).

In simple terms, glacial deposits consist of (in order of increasing age and depth):

- glacio-fluvial outwash sands (LP1) in the northern part of the site;

- several clay-rich units (including, commonly, a layer of clay between 5 and 8 m bgl) (LP2);

- fluvial outwash sands and gravels (LP3);

- a large, laterally continuous thickness of clayey till (LP4);

- tills with occasional meltwater deposits (LP5) – these constitute the majority of superficial deposits (BNFL, 2002d). BNFL (2002d) states that this material lies directly on the sandstone to the north of the site and is absent where the sandstone reaches its highest elevation (northern corner of the site);

- silts and clays associated with marine deposition (LP6) (BNFL, 2002c,d);

- the lowest till layer (LP7) (not always present).

More detailed descriptions of each lithofacies package are given in the following text and are represented in Figure 1.

3.2.3. Lithofacies package description

Lithofacies Package 1 (Late Devensian to Holocene Peat, and other recent deposits)

Lithofacies Package 1 (LP1) is regarded as once forming a continuous 'peat' layer. Hunter *et al.* (2008) notes that the use of the word 'peat' in borehole logs from the site is a loose term covering a thin (1-3 m) succession of laminated clays overlain by fluvial sands and gravels and a layer of peat. This is 'draped' across the site and covered by a varying thickness of surficial dune sand.

Lithofacies Packages 2 and 3 (alternating tills, sands and gravels; incised channel gravels)

Three clayey tills are penetrated by LLWR site boreholes and have been grouped together to form Lithofacies Package 2 (LP2).   These tills comprise part of a sequence of

alternating sands and clayey till lithofacies immediately beneath the peat layer at the LLWR site. This package includes several wide, shallow, linear features possibly representing 'incised channels' that appear to cut down into (but not through) the underlying Lithofacies Package 4 (Thick Till) into Package 5. The gravel infill of these 'channel' features forms Lithofacies Package 3 (LP3) (Hunter *et al*. 2008, updated by Smith, 2008). The features themselves appear to be orientated in a north-east to south-west direction. Geophysical (resistivity) information suggests that the channel features may extend further in the direction of the coast (Smith, 2008).

Lithofacies Package 4 (Thick Till)

The underlying Lithofacies Package 4 (LP4) is found at similar elevations in all the boreholes drilled to the relevant depth within the site and its immediate surroundings. This unit, which represents the Main Diamict described in BNFL (2002c,d) and the regionally-defined Holmrook Till Member of the Central Cumbria Drift Group (Merritt and Auton, 2000), comprises a laterally-continuous, relatively thick, clay-rich till unit, often called the 'Thick Till'. Within the site, the package as a whole dips shallowly to the south-east. There is insufficient information between the site and the coast to describe the slope of this unit in this area.

3D modelling of the lithopackage surfaces (Smith, 2008) suggests that the 'channels' of LP3 incise into (but not through, as initially thought by Hunter *et al*., 2008) LP4. Although these 'channels' are now thought not to penetrate LP4, the latter possesses a laterally-variable lithology, ranging from clay through to silts (Smith, 2008), which may still indicate a possible route for groundwater migration into the underlying sediments.

Lithofacies Package 5 (silts, sands and gravels)

Hunter *et al*. (2008) describe Lithofacies Package 5 (LP5) as silts, sands and gravels occurring beneath the thick clayey layer of LP4. Where Lithofacies Package 6 (LP6) is absent, the base of LP5 is in contact with rockhead. It is also important to note that this LP5 onlaps onto the rockhead high towards the north of the LLWR site, defining its northern boundaries (i.e. the sandstone penetrates through LP5 so that LP4 sits on the rockhead high). LP5 varies in thickness considerably and the lower parts of the package also onlap onto a shallow rockhead high in the southern area of the LLWR.

Lithofacies Package 6 (silt)

This package is a relatively thick silt layer restricted to the south-eastern half of the site. It is likely to pinch out against an embayed, rising bedrock topography. This silt layer is only recorded at four of the cluster borehole locations within the LLWR site and at one offsite borehole, located in the coastal dunes south of the beach access road.

Lithofacies Package 7 (older, thin basal tills, sands and gravels)

Package 7 forms the infill of any voids underlying LP6. It is thus a localised deposit, only present towards the south-eastern area of the LLWR site.
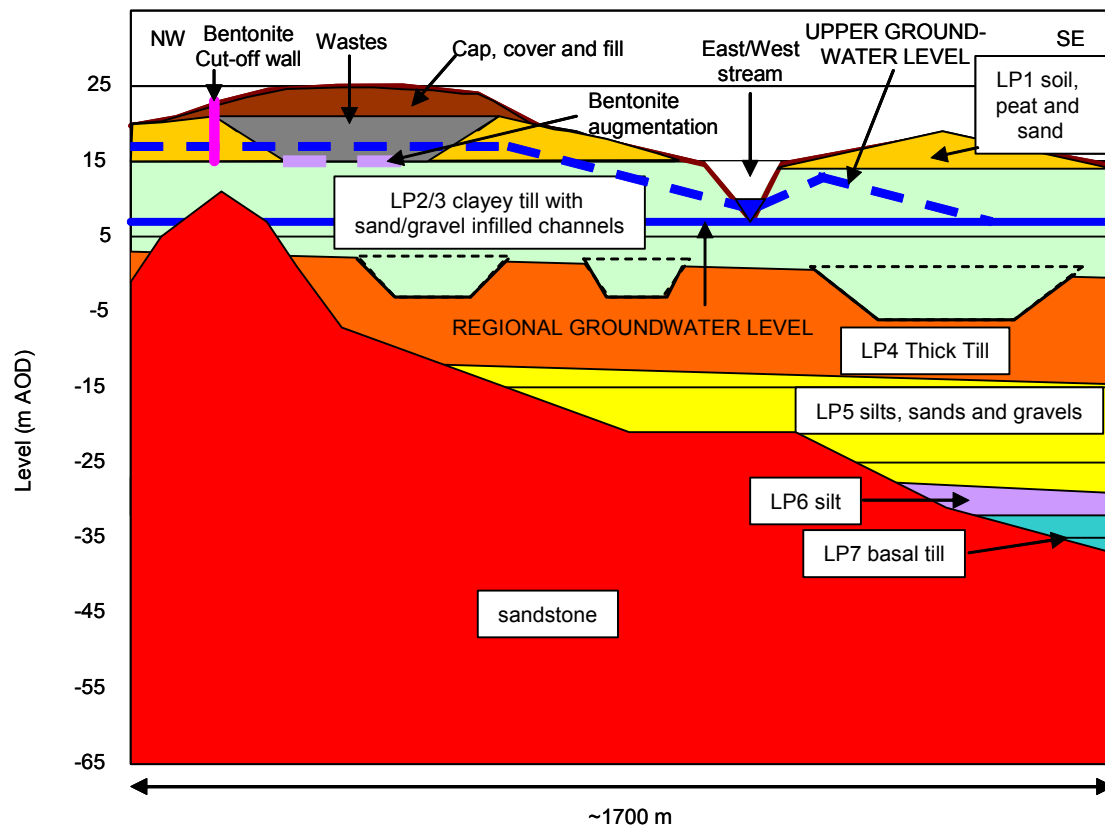
Figure 1: Schematic illustration of lithofacies packages and relative levels of Upper and Regional groundwater (based on information from Hunter *et al.*, 2008; Smith, 2008 and Figure 2 of BNFL, 2002b).

3.3. Mineralogy

The mineralogy of sediments present in the geosphere of the LLWR has been previously examined in a number of studies (e.g. Williams *et al.*, 1985; Bloodworth and Inglethorpe, 1988; Milodowski, 1990; Berry *et al*., 1990; Loftus, 1995), and updated as part of the 2002 PCSC (Randall, 2002; Randall *et al.,* 2004a). Since 2002, only a limited study has been undertaken to characterise the sediments utilised in the uranium sorption experiments presented in Dutton and Trivedi (2008).

The common rock forming silicates, such as quartz, feldspars (plagioclase and potassium feldspar) and micas dominate the bulk mineralogy of the sediments. Quartz is the dominant mineral in coarse size fractions whilst feldspars and mica become more abundant in the clay size fractions. Two carbonate minerals have been detected; dolomite, mainly as a trace component of the finer fractions, and calcite, which has been identified in a number of samples but appears to be concentrated in the shallower, clay-rich horizons.

A complex mix of phyllosilicates is present in the sediments, exhibiting a wide range of types and degrees of order, with detrital and weathering products present. The detrital material is generally coarse grained and well crystallised, consisting of micas and chlorites, including muscovite, biotite and chlorite. These appear to be derived from granites, pegmatites and some metamorphic material. Well crystallised kaolinite is also

occasionally observed. Cleavages have been identified in feldspar, indicating alteration during weathering, resulting in the potential release of cations into solution, the production of significant secondary porosity or the formation of secondary phyllosilicate phases. The secondary phases include illite and poorly ordered swelling clays, the latter identified as vermiculite, formed under near-surface conditions by weathering, hydration and partial alteration of coarse grained detrital mica, in particular biotite (the vermiculite phase contains high potassium, iron and magnesium similar to biotite). These secondary phases are most often found on the surfaces of primary material.

Iron oxide/hydroxide minerals are detected within the sediments, predominantly present as very fine-grained minerals and often associated with the clay coatings. Where grain size is larger, these minerals show a crystal structure indicative of haematite. There is also evidence that, as well as discrete mineral phases, many surfaces are coated with iron oxides. Other iron bearing minerals present are pyrite, chlorite and carbonate minerals, with siderite being identified in samples taken from the clay rich LP2/3 formation.

Table 3 summarises the mineral content of ten LLWR samples taken from various formations for the 2002 PCSC, whilst Table 4 provides similar information for the sediments used for the recent uranium sorption experiments (Dutton and Trivedi, 2008).

In terms of sorption, cation exchange capacity and specific surface areas are perhaps more useful indicators than bulk mineralogy. The data available for these parameters are limited but Figure 2 indicates the relationship between clay content and measured cation exchange capacity from analyses carried out by Milodowski (1990) and the characterisation undertaken in support of the 2002 PCSC (the latter data reproduced in Table 5). There is a clear relationship between clay content and measured cation exchange capacity, although the data from Milodowski (1990) indicate a lower cation exchange capacity for given clay content. The source of these differences is not clear but is most likely related to either size separation techniques or a real difference between sediments taken from different locations. Of some support to the latter explanation is the fact that one of the samples from the 2002 PCSC dataset, a sample from the shallow LP2/3 formation, plots close to the Milodowski data. The samples from Milodowski are relatively shallow, which indicates that they may have come from the LP2/3 formation and may be associated with clay minerals with lower cation exchange capacity. Table 5 indicates that the cation exchange capacity of the clay-sized fraction of these shallow samples is less than that of the deeper samples. This is potentially due to the lower concentrations of the high cation exchange capacity clay phase vermiculite in the clay size fractions for the LP2/3 formation.

Another potential measure of sorption potential is the specific surface area, measured through the Brunauer, Emmett and Teller (BET) technique. Figure 2 shows the comparison between the measured clay and specific surface area. As can be seen from the graph, the relationship between clay content and surface area is strong and the difference between the LP2/3 samples and other formations is not apparent, in contrast to the cation exchange capacity data.

Table 3: Semi-quantitative mineral description of LLWR sediment samples.

Bulk samples (top) and clay minerals in the <2µm size fraction (bottom)

Borehole	Depth m	Formation	Quartz %	K-feldspar %	Plagioclase %	Siderite %	Calcite %	Amphibole %	Di-clay * %	Tri-clay ** %
6123	4.5-4.95	LP2/3	47.4	9.2	6.8	0.7	1.9	-	25.2	8.8
6124	6.8	LP2/3	46.8	9.3	6.2	0.8	1.2	-	25.4	9.3
6125	13.8	LP3	74.0	6.8	5.8	-	-	-	7.4	6.1
6123	22	LP4	48.8	13.7	16.7	-	-	trace?	10.9	9.9
6123	22.5	LP4	41.0	11.6	17.0	-	-	trace?	19.0	11.4
6125	25.2	LP4	47.0	9.5	13.9	-	-	-	18.2	11.4
6124	27-27.5	LP4	52.7	13.7	15.6	-	-	1.9	5.7	10.4
6125	28	LP4	38.0	11.3	23.6	-	-	3.0	10.5	13.6
6124	31	LP4	62.7	10.0	8.9	-	-	-	12.1	6.2
6123A	38	Sandstone	83.8	8.8	1.9	-	-	-	5.6	-

*Total dioctahedral clays   **Total trioctahedral clays

Borehole	Depth m	Formation	Illite %	Chlorite %	Chlorite-vermiculite %	Vermiculite %	Smectite %	Kaolinite %
6123	4.5-4.95	LP2/3	67	7	10	1	-	15
6124	6.8	LP2/3	63	5	20	1	-	11
6125	13.8	LP3	69	2	16	5	-	8
6123	22	LP4	57	5	31	4	-	3
6123	22.5	LP4	55	1	26	14	-	4
6125	25.2	LP4	61	5	19	8	-	7
6124	27-27.5	LP4	64	5	17	11	-	3
6125	28	LP4	58	8	26	3	-	5
6124	31	LP4	72	1	16	4	-	7
6123A	38	Sandstone	70	1	-	-	28	1

Table 4: Semi-quantitative mineral description of LLWR sediment samples (Dutton and Trivedi, 2008).

Bulk samples (top) and clay minerals in the <2µm size fraction (bottom)

Borehole	Quartz %	K-feldspar %	Albite %	Calcite %	Chlorite %	Dolomite %	Haematite %	"kaolin" %
Clay sediment 1	51.2	7.0	6.8	2.6	7.7	2.9	0.7	4
Sand sediment 1	82.0	1.2	6.5	nd	1.8	nd	<0.5	0.6

Sample	Illite %	Chlorite %	Chlorite/smectite %	Kaolinite %	Surface Area $m^2 g^{-1}$	CEC meq $100g^{-1}$
Clay sediment 1	76	11	4	9	15.81	18.6
Sand sediment 1	72	13	2	13	1.83	8.0

Table 5: Cation-exchange capacity and surface area measurements for LLWR sediments.

Note: Cation exchange capacity (CEC) shown for both the bulk sediment and the clay-size fraction (<2µm)

Borehole	Depth m	Formation	CEC (bulk) meq $100g^{-1}$	Surface Area $m^2 g^{-1}$	CEC (clay) meq $100g^{-1}$
6123	4.5	LP2/3	15	11.3	18.3
6124	6.8	LP2/3	5.4	12.9	19.4
6125	13.8	LP3	4.1	4.0	24.2
6123	22	LP4	4.7	1.6	27.4
6123	22.5	LP4	13.9	10.5	33.6
6125	25.2	LP4	12.2	13.1	31.9
6124	27	LP4	n/d	0.8	28.4
6125	28	LP4	n/d	1.0	22.6
6124	31	LP4	7.5	6.9	30.9
6123	38	Sandstone	<3	1.6	32.4
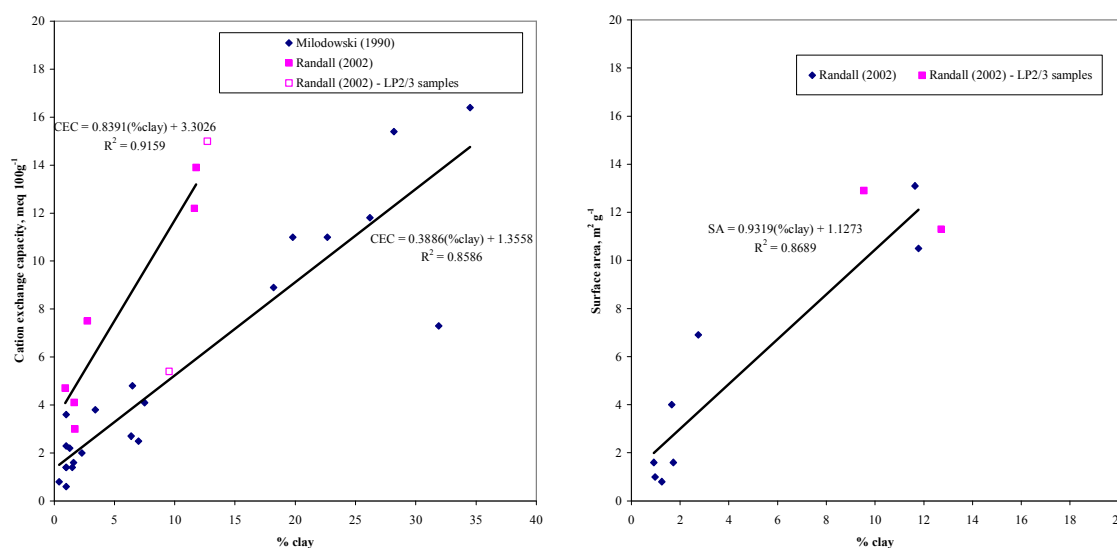
Figure 2: Relationship between clay content and cation exchange capacity (left) and surface area (right).

From the information provided in Table 5 and Figure 2, it is clear that, for the data obtained during the 2002 PCSC (which represent a greater depth profile than previous data), the sediments split into two broad groups. Firstly, there is a group of coarse sediments with a clay content of <5% and associated cation exchange capacities of between 3 and 7 meq $100g^{-1}$ and surface areas of between 1 and 4 m^2 g^{-1}. A second group represents silt sediments, with clay content of around 12% and associated cation exchange capacities of between 12 and 14 meq $100g^{-1}$ and surface areas of between 10 and 13 m^2 g^{-1}. These distinctions will be used in the specification of K_ds presented in the later sections of this report.

3.4. *Groundwater chemistry*

The following information is a summary of previously reported work (BNFL, 2002e). LLWR groundwater has a predominantly fresh water composition and is calcium bicarbonate dominated. LLWR groundwater samples are either slightly undersaturated or close to equilibrium with calcite, which controls the concentration of calcium and bicarbonate ions and also the pH. Partial pressures of CO_2 are elevated with respect to atmospheric composition.

The importance of meteoric water on the composition of the LLWR groundwater has been demonstrated (Section 5.2, BNFL, 2002e), with a concentration factor of approximately three required to account for the concentrations of sodium, chloride and sulphate in groundwater. Stable isotope data also indicate that meteoric recharge is the main process controlling δD and $\delta^{18}O$ composition, with isotope exchange resulting from the geochemical processes being minimal.

Enhancement of magnesium in LLWR groundwater is due to carbonate and ion exchange reactions, consistent with mineralogical evidence (Section 5.3, BNFL, 2002e). Similarly, potassium concentrations in groundwater are enhanced with respect to the evaporated rainwater model due to ion exchange equilibria, due to clay minerals with a high affinity for potassium being active in the LLWR geosphere. This is consistent with mineralogical

information, where illite and vermiculite dominate the clay assemblage, formed by the weathering of coarse grained, high-temperature phases such as feldspars and micas. Weathering of these minerals also releases silica and aluminium into solution and secondary weathering products.

LLWR groundwaters may be classified as moderately reducing (Section 5.4, BNFL, 2002e). Manganese concentrations indicate that all manganese minerals are undersaturated, consistent with conditions of manganese reduction. Redox conditions appear to be controlled by iron minerals and can be represented by considering equilibrium between iron oxyhydroxides and siderite, both of which have been identified in LLWR sediments.

A "reference groundwater" has previously been specified, which has been used as the basis of the aqueous phase composition for site specific sorption experiments. Table 6 shows the composition of this reference groundwater (see Randall, 2002).

The speciation of redox sensitive radionuclides included in the current radiological assessment has also been determined for this reference groundwater and is shown in Table 7.

Table 6: Composition of reference groundwater.

	Concentration, mol l^{-1}
pH	7.349
pe	2.868
Ca	2.012e-3
Mg	7.942e-4
Na + K	1.359e-3
Fe	1.185e-5
Si	1.608e-4
Cl	1.340e-3
CO_3^{--}	5.365e-3
SO_4^{--}	3.958e-4
NO_3^{-}	1.100e-4

Table 7: Calculated speciation of redox sensitive radionuclides under reference groundwater conditions.

Element	Oxidation State	Speciation	
Neptunium	73% as Np(V)	NpO_2^{+}	60%
	27% as Np(IV)	$Np(OH)_4$	27%
		$NpO_2CO_3^{-}$	13%
Protactinium	100% as Pa(V)	$PaO_2(OH)_2$	100%
Plutonium	100% as Pu(IV)	$Pu(CO_3)_3^{2-}$	99%
Technetium	100% as Tc(VII)	TcO_4^{-}	100%
Uranium	100% as U(VI)	$UO_2(CO_3)_2^{2-}$	53%
		$UO_2(CO_3)_3^{4-}$	45%

4.   Sorption database

4.1.   Introduction

The sorption parameters presented in this report are based on those derived by Randall (2002), which utilised a range of site specific experimental studies (listed in Appendix A). Appendix A also gives details of the groundwater compositions used in these experiments and also the key features of the experimental protocols.

This dataset was carefully screened by Randall (2002) to ensure that only those experiments carried out under conditions representative of the LLWR site were used. All experiments used only real LLWR sediments rather than simulants or analogues. The aqueous phase used in the experiments was either groundwater obtained from boreholes at the site or a simulated groundwater appropriate to conditions in the LLWR geosphere.

For the majority of the experiments no attempt was made to control the pH, with the system allowed to reach an equilibrium pH. Exceptions to this include Yule (1994), who systematically varied pH between 6 and 8, and Berry *et al.* (1990). These variations were achieved through addition of acid or base to the starting solution. Similarly, no attempt was made in any experiment to control redox conditions. All experiments were exposed to air and thus aerobic. This would be potentially problematic if the data were used to construct a sorption database for a site associated with anaerobic conditions. However, the LLWR geosphere is characterised as moderately reducing and Table 7 reveals that redox sensitive elements, such as uranium and technetium, are present in their higher oxidation states. Additionally, the detailed speciation modelling of all experimental systems presented in Randall (2002) also shows that the redox states of radionuclides are consistent with those predicted to dominate in the geosphere. Finally, none of the experimental studies involved control of pCO_2. All experiments were carried out under atmospheric pCO_2, which is lower than that encountered in the LLWR geosphere (Section 3.4). For some radionuclides, such as uranium, complexation with dissolved CO_2 can reduce sorption and shift the sorption maximum to lower pHs. The most recent uranium experiments (Section 4.11) were carried out at 1% CO_2, which is more appropriate to the observed LLWR conditions. These experiments demonstrate that higher levels of pCO_2 reduce uranium sorption at neutral or higher pH values and so it is recommended that any future experiments be carried out under these conditions.

Appendix A also shows the key features of the experimental methodologies. Although some of the studies were performed over 20 years ago, in general, the methodologies are considered reasonable compared to today's standards. For example, four of the experimental studies used 30000 MWCO filters for phase separation, which is consistent with best practice today (and is the approach used in the most recent uranium studies presented in Section 4.11).

The experimental protocols have been re-evaluated since the production of Randall (2002). For a number of experimental determinations of K_ds, the experimental protocol has been identified as suspect or problematic (Randall *et al.*, 2004b), and these have been removed from consideration. These studies include Stafford (1993, 1995) where poor control of the pH prior to the addition of radionuclides is likely to have led to polymerisation of some elements, particularly the actinides. Therefore, the measurements from these sources for uranium, neptunium, americium and plutonium

have been removed from the K_d calculations. However, the data for radium from these sources have been retained as the likelihood for polymerisation is deemed low. Additionally, some data from Berry *et al.* (1990) have been removed where the experimental methodology cannot be verified. This is particularly at high mass:volume ratios, where poor mixing is likely to have led to artificially high K_ds.

Thus, given the screening process presented in Randall (2002) and the subsequent evaluation described above, it is considered that the experimental systems used in the construction of the sorption database are robust and representative of the environment associated with the LLWR geosphere.

Secondly, it has been decided to restrict the pH range for elements that demonstrate high dependence for sorption. As shown in BNFL (2002e), the average pH for the LLWR groundwater is 7.15 ± 0.55 on the basis of post-completion borehole sampling. Therefore, only sorption data from experiments carried out between approximately pH 7 – 8 have been considered.

Although there is significant heterogeneity in sediment texture present at the LLWR site (ranging from clays to gravel-rich horizons) it is deemed appropriate to utilise the "coarse" classification, as was used in the 2002 PCSC (Randall, 2002), to represent the drift sediments. The discussion presented in Section 3.3 indicates that the Thick Till formation (LP4) is dominated by relatively coarse sediments. The results of experiments for sorption onto silt sediments, however, do provide an upper bound for any future sensitivity studies for the groundwater pathway assessment.

Representation of the sandstone is more problematic as no site specific measurements exist. For the 2002 PCSC, data from Quantisci (1999) were used. Data for the region are available from the Nirex programme (Nirex, 2003) for sorption to sandstone. However, it is not clear whether the groundwater conditions are appropriate to the shallow regions of the sandstone relevant to the groundwater pathway within the current radiological assessment models.

Therefore, sandstone K_ds have generally been estimated through consideration of the mineralogical properties. As shown in Randall (2002), only one sandstone sample has been characterised with resultant cation exchange capacity (CEC) of < 3 meq 100g^{-1} and a specific surface area of 1.6 m^2 g^{-1}. This is compared to an approximate CEC for coarse drift sediments of $\sim$ 5 meq 100g^{-1} and a surface area of between 1 and 7 m^2 g^{-1}. Therefore, in deriving appropriate K_ds for the sandstone, the drift sediment K_ds have been adjusted by a factor of approximately 0.6 – 0.75 depending on the radioelement.
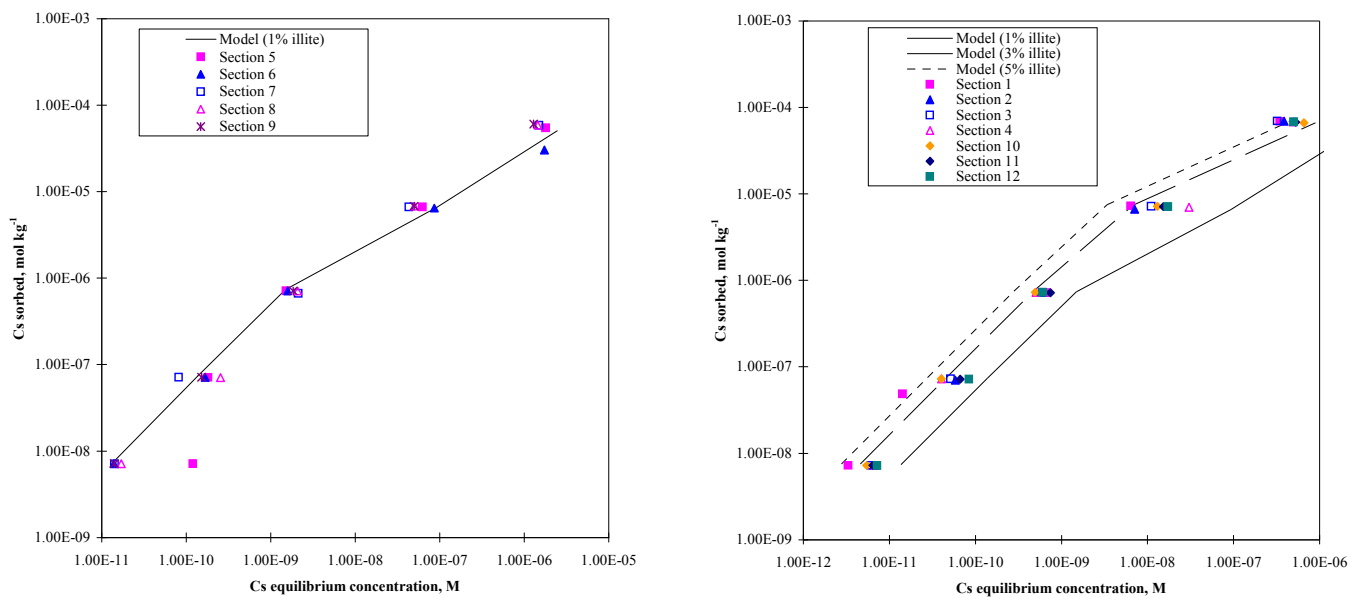
4.2. Caesium

4.2.1. Geochemistry

The solubility of caesium is very high and so the main process controlling the mobility and distribution of caesium in the environment is sorption. The mobility of caesium has been the subject of extensive investigation over many years due to its introduction to the environment as a result of nuclear weapons testing and nuclear accidents, such as Chernobyl. These studies have indicated the strong interaction between caesium and solid phases. The principal mechanism for caesium sorption onto geological materials is generally accepted as being cation exchange but with a particular affinity for micaceous minerals, such as illite (e.g. Comans *et al*., 1991; de Preter *et al*., 1991; Sawhney, 1972). This affinity arises because of the similarity of caesium to potassium, in terms of low hydration energies, allowing the ion to fit between layers of the micaceous minerals, whereas larger cations such as calcium are excluded due to their size. The high affinity of micaceous minerals for caesium means that the extent of sorption may be considerably higher than would be indicated by a simple cation exchange capacity value.

A strong dependence of concentration on sorption has been demonstrated by several studies onto different media (de Preter *et al*., 1991; Sawhney, 1972). In terms of sorption onto sediments encountered at the LLWR site, the most comprehensive set of data comes from Haigh and Hooker (1988), which examined sorption for various caesium concentrations onto 12 sub-samples from the site, with varying particle size distributions and cation exchange capacities. As discussed in Randall (2002), modelling these experiments through the use of standard cation exchange constants (Appelo and Postma, 1993) resulted in predicted sorption several orders of magnitude below the measured value. This is in contrast to the good fit between model results and experimental results in the case of strontium. Instead, a more complex model involving using the approach developed by Bradbury and Baeyens (2000) has been applied. Here the sorption of caesium is simulated through a three-site cation exchange model, with the assumption that sorption is dominated by the illite component of a sediment. The three sites represent frayed edge, Type II and planar sites of illite, with selectivity coefficients for caesium and other groundwater cations determined from experiments carried out with a "reference illite".

Figure 3 shows the calculated sorption isotherms for the two sediment textures (sand and silt) studied by Haigh and Hooker (1988). The calculated isotherm, assuming a total of 1% illite, is an excellent fit to the experimental data for the sand samples, whereas an illite content of 3% is a better description of the silt isotherm. These illite concentrations are consistent with semi-quantitative XRD determinations described earlier, where illite makes up approximately 60 – 65% of the clay size fraction.

Representing these data in terms of K_ds shows clearly the relationship between caesium concentration and sorption (Figure 4). Caesium sorption is linear for initial caesium concentrations up to and including 0.01 ppm. Therefore, the K_ds determined within Randall (2002) only considered experimental data where caesium concentrations were 0.01 ppm or less.

Figure 3: Comparison of modelled isotherm (three site model) of caesium with experimental data for LLWR sands (left) and silts (right).
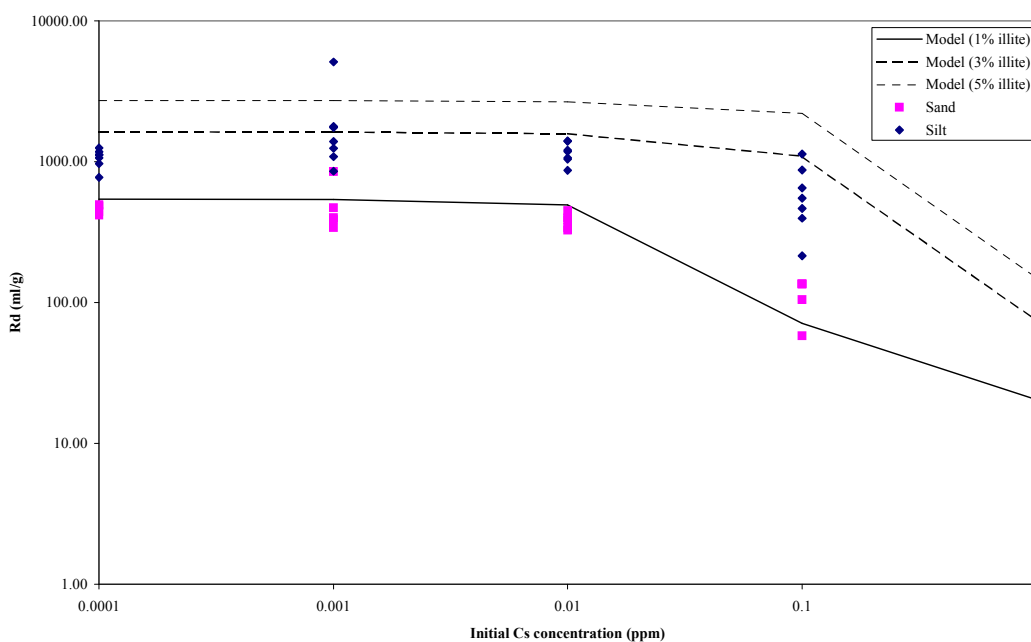
Figure 4: Comparison of modelled K_d as a function of caesium concentration with experimental data.

This model was extended within Randall (2002) to consider ranges of illite concentration that may be encountered in the geosphere, with a resulting simple relationship between K_d and illite content obtained. Figure 5 shows this relationship as a function of both illite content and potassium concentration, the cation that competes more effectively with caesium for sorption sites. The results of this modelling exercise indicate that, for low concentrations of caesium, the K_d can be expressed as a simple function of the illite concentration (Figure 5). If it is assumed that illite makes up approximately 60% of the clay-size fraction, this expression can be further expressed as a function of the clay content, thus:

$K_d = 0.25 \times \%$ clay

Relating this relationship to the classifications of LLWR sediments, Section 3.3 indicates that clay content for representative samples varies from 0.5% to approximately 15%. This results in a range of K_ds from 0.125 m^3 kg^{-1} through to 3.25 m^3 kg^{-1}. For a coarse sediment of approximately 1% clay, a value of 0.25 m^3 kg^{-1} is calculated. Comparing these values to the overall experimental data (see later) reveals a good agreement.
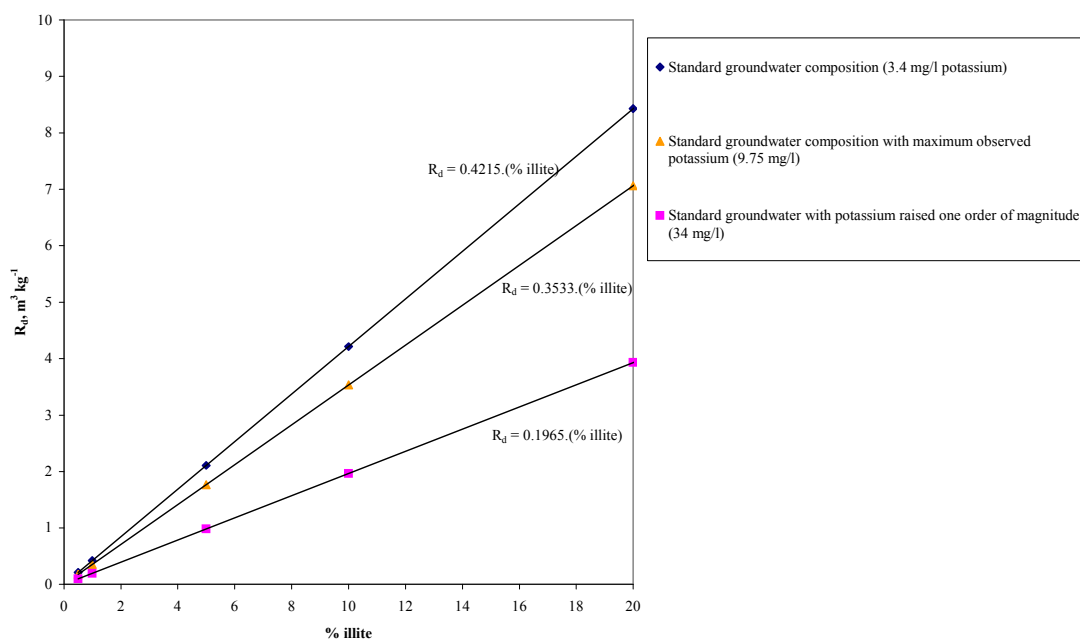
Figure 5: Calculated K$_d$ of caesium as a function of illite concentration and potassium concentration.

4.2.2. Recommended sorption parameters

Within the 2002 PCSC, the sorption of caesium was calculated through consideration of all data for coarse sediments from Yule (1994), Haigh *et al.* (1989), one single datum point from Bell and Bates (1987) (LLWR soil) and certain data (below 0.01 ppm) from Haigh and Hooker (1998). Consideration of these data results in a calculated geometric mean of 0.30 m^3 kg^{-1}.

This is well represented through the ion exchange model described earlier. Given the good agreement between this fundamental modelling and experimental results, it is proposed to keep the value of 0.3 m^3 kg^{-1}.

For silt data, the average from Haigh and Hooker (1988) is 1.2 m^3 kg^{-1}, which is consistent with the range of calculated K$_d$s presented in the previous section.

For the sandstone, a value of 0.1 m^3 kg^{-1} was also used previously. Whilst it may be expected that the sorption onto sandstone will be lower than onto the drift, given the lower CEC and generally lower clay content, in the case of caesium this conclusion is not so straightforward. From the mineralogical analysis presented in Table 3, the illite concentration in the clay fraction of the sandstone is higher than that in the drift. Therefore, it is reasonable to assume that this higher concentration of this mineral phase, which controls caesium sorption, will compensate to some extent the lower clay content. Thus, in the absence of site specific data, it is proposed to use 0.3 m^3 kg^{-1} for the sandstone.

4.3. Strontium

4.3.1. Geochemistry

As an alkali earth metal, strontium exists primarily as the Sr^{2+} ion within the LLWR geosphere groundwaters, with minor contributions from SrSO$_4^0$ (Randall, 2002). The main sorption reaction would be expected to be cation exchange (e.g. Stenhouse, 1995; Grütter *et al.*, 1994; Torstenfelt *et al.*, 1982) and this was confirmed by the detailed analysis of the LLWR site specific data presented in Randall (2002).In particular, the experiments of Haigh and Hooker (1988) were modelled through application of an ion-exchange model by assuming that the measured cation exchange capacity is the main indicator of the extent of sorption. The results are shown in Figure 6, where the cation exchange capacities range from 2.1 to 5.1 meq 100g^{-1}. Excellent agreement is seen between modelled and experimental K$_d$s, indicating the sorption of strontium onto LLWR sediments is controlled by cation exchange reactions. This conclusion is also supported by an experimental study undertaken by Prastka (1999) which revealed significant decreases in sorption as ionic strength increased; consistent with a cation exchange reaction.
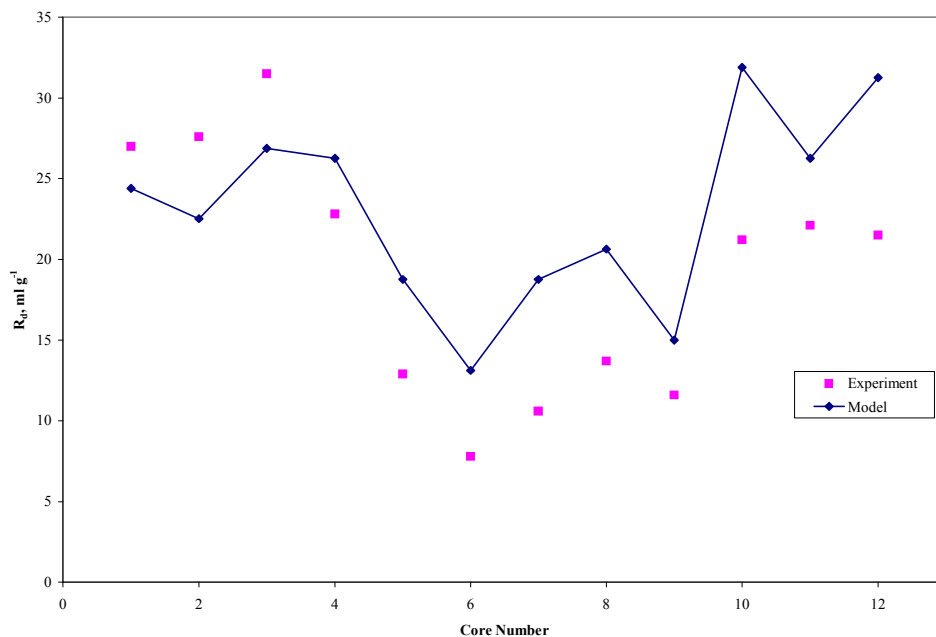
Figure 6: Comparison of modelled strontium sorption, using a simple ion exchange model, and experimental data (Haigh and Hooker, 1988).

Given this conclusion, the ion exchange model can be simply extended to investigate variations is strontium sorption as a function of measured cation exchange capacities and calcium concentration. Figure 7 shows the variation of modelled K_d with a variation of cation exchange capacity from 0.01 to 20 meq $100g^{-1}$, under groundwater conditions defined by Table 6 (*i.e.* with a calcium concentration of 2.012E-03 mol l^{-1}). The result is a linear relationship, thus:

K_d (m^3 kg^{-1}) = 0.0025 $\times$ CEC (meq $100g^{-1}$)

Figure 8 shows the predicted variation in strontium K_d as a result of altering the calcium concentration from 2 – 178 mg l^{-1}, representing the spread of data as reported in BNFL (2002c). From these results, the highest K_d values are predicted at the lowest calcium concentrations and the highest cation exchange capacities. The predicted K_d values range from 0.00013 to 0.148 m^3 kg^{-1}, representing sorption onto a variety of substrates from coarse to clay-rich. The effect of increasing calcium concentration from 2 to 178 mg l^{-1} is to reduce the K_d by a factor of approximately 7.
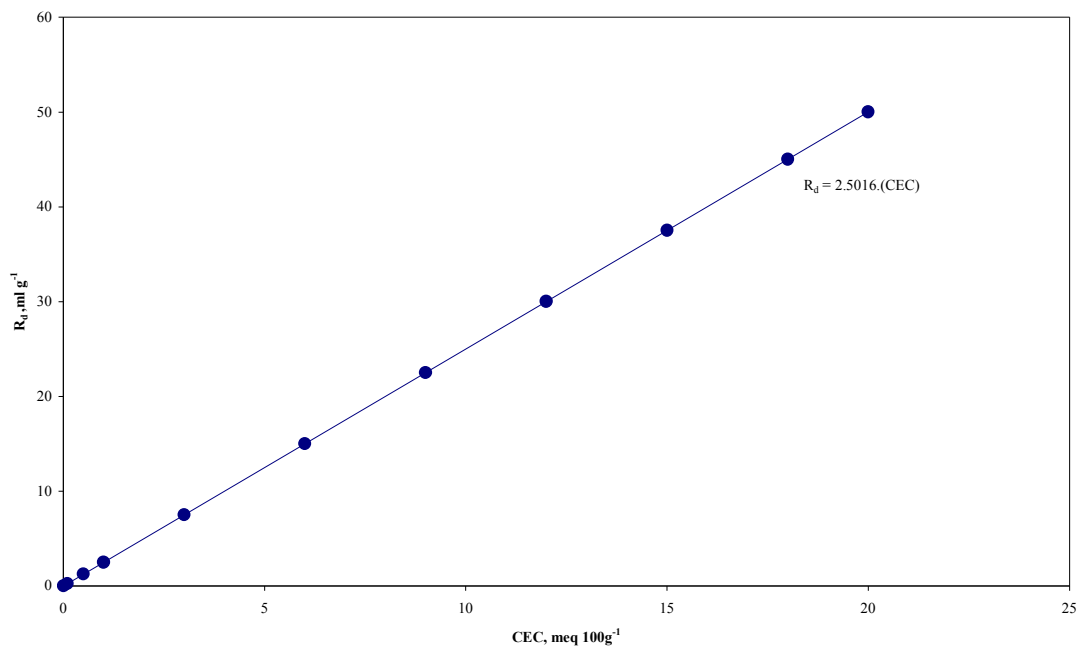
Figure 7: Modelled strontium sorption, using a simple cation exchange model, as a function of cation exchange capacity.
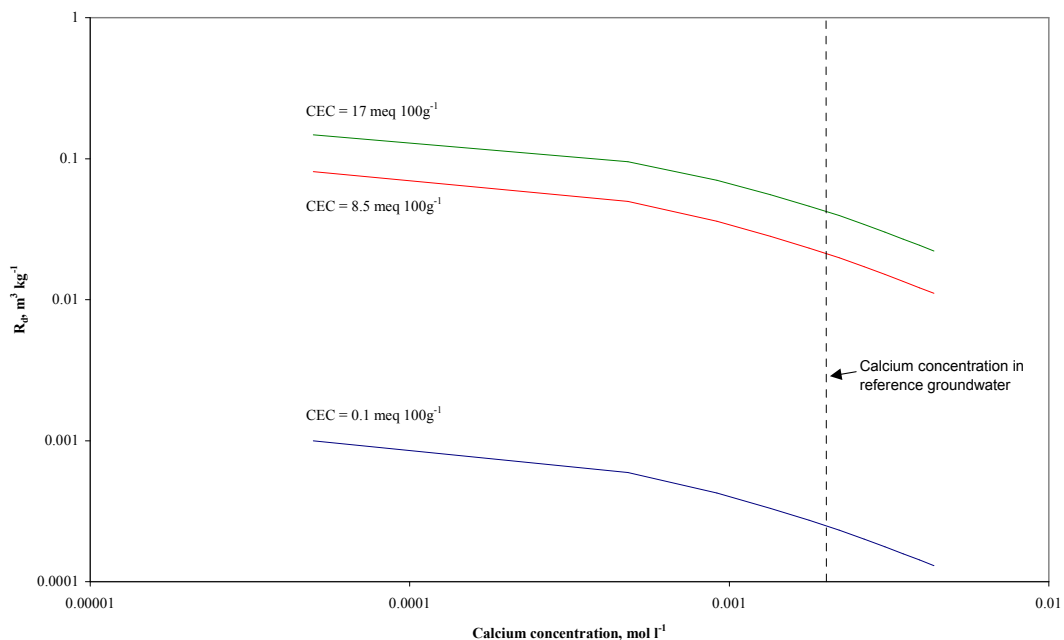
Figure 8: Modelled strontium sorption, using a simple ion exchange model, as a function of cation exchange capacity and calcium concentrations.

4.3.2. Recommended sorption parameters

The discussions within Randall (2002) indicate a geometric mean K_d of 0.012 m^3 kg^{-1} for coarse sediments, with a range from 0.0078 to 0.023 m^3 kg^{-1}. For silt sediments, the geometric mean is equal to 0.02 m^3 kg^{-1}, with a range from 0.008 to 0.032 m^3 kg^{-1}.

These results are consistent with the sorption modelling presented above, particularly the linear relationship between K_d and cation exchange capacity. Using this expression and an assumed average cation exchange capacity of 5 meq $100g^{-1}$ for coarse sediments results in a calculated K_d of 0.0125 m^3 kg^{-1}, almost identical to the average obtained from site specific experiments. Similarly for silt sediments with a cation exchange capacity of approximately 12 meq $100g^{-1}$, a value of 0.03 m^3 kg^{-1} is obtained, which is again within the range of experimental measurements.

For the sandstone, a value of 0.3 m^3 kg^{-1} was used previously. Given that the bulk CEC of the sandstone is lower than the drift and the sorption of strontium is related to the CEC, it is inappropriate to use a higher K_d in the sandstone. The CEC for the only Sandstone sample characterised is <3 meq $100g^{-1}$, whereas the coarse drift sediments are approximately 5 meq $100g^{-1}$. Using the expression derived earlier relating K_d to cation exchange capacity results in a recommended K_d of 0.0075 m^3 kg^{-1}, a reduction by a factor of 0.6 compared to the drift value.

4.4. Radium

4.4.1. Geochemistry

Radium is an alkaline earth metal and behaves in a similar way in aqueous solution to calcium and strontium, in that Ra^{2+} dominates its speciation. Speciation of the groundwaters used in the site specific sorption experiments indicates that Ra^{2+} is predicted to be the dominant species, with some contribution from $RaSO_4^0$ (Randall, 2002): up to 33% of the speciation in Braithwaite (1997) although sulphate concentration was very high in these experiments (10 mM). The other site specific experiment (Stafford, 1995) used a groundwater with 0.1 mM, with a consequent lower contribution from $RaSO_4^0$ (5%) (Randall, 2002).

The sorption of radium is expected to be dominated by ion exchange reactions, with consequent dependence on ionic strength, cation exchange capacity and the presence of competing ions such as calcium and magnesium.

Given the shortage of site specific experiments, it is impossible to examine the processes governing sorption onto LLWR sediments. The work of Stafford (1995), however, indicates little change in sorption behaviour due to the presence of natural organics.

4.4.2. Recommended sorption parameters

For coarse sediments, only data from Stafford (1995) were used in the 2002 PCSC. Although there are questions regarding the experimental technique, it is assumed that this is more of an issue for actinides than for radium, which does not hydrolyse to the same extent. Therefore, the K_d of 0.086 m^3 kg^{-1} is kept for the current radiological assessment.

Just one single datum point is available for sorption onto silty sediments - a site specific K_d of 0.057 m^3 kg^{-1} (Braithwaite (1999). There is very little difference between the site specific K_ds for coarse sediments and silty sediments, although as noted above sulphate concentrations were very high for this experiment, potentially reducing sorption.

For the sandstone, a value of 0.3 m^3 kg^{-1} was used previously. As for the case of strontium, the sorption of radium is expected to the dependent on the CEC of the sediment, and so the same arguments are valid for the derivation of a more appropriate K_d (i.e. a reduction by a factor of 0.6). This results in a K_d of 0.05 m^3 kg^{-1}.

4.5. Nickel

4.5.1. Geochemistry

Aqueous nickel chemistry is dominated by that of Ni(II). It is expected to form a wide range of aqueous species particularly carbonates and hydroxides. Speciation calculations indicate that $NiCO_3$ is the dominant species under the conditions examined in site specific experiments, with Ni^{2+} becoming more important at lower pH.

Sorption is likely to be controlled by surface complexation reactions. Randall (2002) reports on the experiments of Prastka (1999), which showed minimal influence of ionic strength on sorption. This indicates that cation exchange is not a dominant mechanism. Similarly, some variability in sorption as a function of pH is observed in two site specific studies (Polfer and Braithwaite (1998); Yule (1994)). This is indicative of a surface complexation reaction.

4.5.2. Recommended sorption parameters

For the 2002 PCSC, the results of Polfer and Braithwaite (1998) and Yule (1994) were used in deriving a K_d. However, this included data at both pH 6 and pH 8, which is deemed inappropriate. Therefore, only one value from Yule (1994) measured at pH 7 is used: 0.04 m^3 kg^{-1}.

For the sandstone, a value of 0.01 m^3 kg^{-1} was used previously. However it is deemed appropriate to use a scaling factor based on the lower surface area of the sandstone. Applying a factor of 0.7 results in a K_d of 0.03 m^3 kg^{-1}.

4.6. Cobalt

4.6.1. Geochemistry

The aqueous chemistry of cobalt is dominated by Co^{2+} and it is expected to form both organic and inorganic complexes in natural groundwaters.

4.6.2. Recommended sorption parameters

Cobalt was not included in the 2002 PCSC although some site specific sorption data are available: Haigh *et al.* (1989) and McKinley and West (1981). For the latter, pH is not given and so data are taken from Haigh *et al.* (1989), with a K_d of 0.14 m^3 kg^{-1}. Applying a scaling factor of 0.7 to account for the lower surface area of the sandstone samples gives a recommended K_d for sandstone of 0.1 m^3 kg^{-1}.

4.7. Technetium

4.7.1. Geochemistry

Technetium can exist in all oxidation states from +7 to −1, although +7 and +4 are the more stable in solution. Tc(VII), as the TcO_4^- ion, is the predominant species under oxidising conditions, and is highly mobile, exhibiting little sorption or precipitation. Under reducing conditions, Tc(IV) becomes predominant, with a consequent lower mobility.

Randall (2002) indicates that, under the reference groundwater conditions indicated in Table 6 , technetium will be present as Tc(VII). Similarly, both the site specific sorption experiments listed in Randall (2002) were carried out under oxidising conditions and so the technetium is also present as TcO_4^-.

Sorption of technetium under oxidising conditions is very low (e.g. see the survey of technetium sorption determinations presented in Stenhouse, 1995). In terms of site specific experiments, Yule (1994) looked at the effect of pH and humic acid on the sorption of technetium onto sand. It is difficult to draw any conclusions from this work, as the K_d values are extremely low, even apparently negative in some cases (representing an insignificant difference between initial and final concentrations). The measured K_d values are only non-negative when humic acids are present, but even in this circumstance, K_d values are low (0.001 m^3 kg^{-1}).

The low sorption shown in the site specific experiments is not surprising, given the speciation of technetium as an uncomplexed anion. The presence of organic material in the sediment will likely have a large effect on the sorption of technetium – Henrion *et al.* (1985) noted non-zero sorption onto Boom Clay, which is probably attributable to the reduction of technetium to Tc(IV).

4.7.2. Recommended sorption parameters

There are two site specific data sources (Yule, 1994; Williams *et al.*, 1985) reported in Randall (2002), both of which indicate very low sorption. It is therefore appropriate to assume a K_d of 1E-04 m^3 kg^{-1} to represent sorption onto all sorption substrates.

4.8. Carbon

4.8.1. Geochemistry

The chemistry of C-14 in the geosphere will be complicated by the amount of non-radioactive carbon naturally present. Significant amounts of calcite and other carbonate minerals are present in the LLWR, with the groundwater chemistry being controlled, to a large extent by the equilibrium with these minerals. Therefore, the retention mechanism is likely to be a complex mixture of isotope exchange and precipitation.

4.8.2. Recommended sorption parameters

No site specific data were available for the derivation of K_ds within the 2002 PCSC, and the same is true in 2008. Within the 2002 PCSC, a K_d of zero was assumed based on the lack of data. However, carbon is a reactive element within the LLWR geosphere, with calcite present in sediments, and groundwater conditions are largely controlled by the equilibrium with CO_2. Therefore, some retardation of carbon-14 can be expected, although this is difficult to quantify, given the complex variety of precipitation, isotope

exchange and sorption reactions expected to occur. Thus a small, but finite, value is proposed for the K_d for C-14: 0.0005 m³ kg⁻¹, taken from an experimental study of a similar sediment to that found in the LLWR geosphere (Plummer *et al.*, 2004). This experimental study, whilst not explicitly attempting to differentiate between sorption and precipitation, does represent a detailed investigation of the behaviour of C-14 in the geosphere environment. Large scale column (2.9m long) experiments were carried out to investigate both gas phase and aqueous phase transport, with K_ds calculated from breakthrough curves. For this report, K_d values from these breakthrough curves were used rather than the K_ds determined from soil sample analysis undertaken after one year. This was undertaken to ensure that the data are more appropriate to a relatively rapid flowing system, with sorption dominating, rather than more long term mineralisation and sorption processes. It is therefore considered appropriate to use these data within LLWR assessments, with the K_d values consistent with other safety cases (e.g. SKB, 2004; Thompson and Miller, 2005; Nagra, 2002 and Nirex, 2003).

It is proposed that the value of 0.0005 m³ kg⁻¹ should also be used for the sandstone.

4.9. Iodine

4.9.1. Geochemistry

Iodine exists in aqueous solution as iodide (I⁻) or iodate (IO₃⁻), the latter more prevalent under highly oxidised conditions. Speciation calculations using the groundwater reported in Haigh *et al.* (1989) confirmed that all of the iodine is present as I⁻.

Little site specific data are available, and so inferences as to sorption mechanisms and controlling factors are difficult. Important parameters that have been identified by other authors include pH (Rancon, 1988), the presence of metal ions that form low solubility iodine solid phases (Strickert *et al.*, 1980) and, perhaps most importantly, the presence of organic material in the sediment. The only conclusions from site specific sorption experiments comes from Haigh *et al.* (1989), who examined the effect of humic acid material in the aqueous phase but found no effect on sorption.

4.9.2. Recommended sorption parameters

There are two sources of site specific data for sorption onto site specific coarse sediments (Williams *et al.*, 1985; Haigh *et al.*, 1989). Both sources indicate low sorption: 0.002 and 0.0006 m³ kg⁻¹ with a geometric mean of 0.0011 m³ kg⁻¹. It is assumed that sorption of the anionic iodide is controlled by the presence of organic material in sediment. It is further assumed that the organic content of the sandstone is minimal and so iodine will not sorb to any significant extent in the sandstone, with a consequent K_d of zero.

4.10. Thorium

4.10.1.Geochemistry

The aqueous geochemistry of thorium is dominated by the Th(IV) oxidation state. Thorium is characterised by its ease of hydrolysis and Th⁴⁺ exists only in very acidic solutions. The aqueous speciation of thorium would be expected to be dominated by

$Th(OH)_4$, and this is confirmed by speciation calculations of the conditions used in the site specific sorption experiments (Randall, 2002).

The very low solubility of thorium in natural waters means that experimental determination of thorium is difficult, with many apparently high K_d values reported in the literature attributable to precipitation of thorium oxides (USGS, 2001).

In terms of site specific experiments, Stafford (1995) noted that the sorption of thorium onto "G6 sand" was not dependent on ionic strength or carbonate concentration. The former suggests that ion exchange is not an important sorption mechanism, at least for sandy sediments (although there is some questions surrounding the experimental protocol used in these experiments). Similarly, LaFlamme and Murray (1987) found that ionic strength did not affect the adsorption of thorium onto goethite.

4.10.2. Recommended sorption parameters

For the 2002 PCSC, only the results of Stafford (1995) and Bell and Bates (1987) were used in deriving a K_d. However, given that there are questions regarding the experimental methodology of Stafford (1995), only the measured K_d from Bell and Bates (1987) is deemed appropriate. This gives a K_d of 0.88 m^3 kg^{-1}.

From Randall (2002), no appropriate site specific sorption experiments were available for silt sediments, with the data from Haigh and Hooker (1988) and Braithwaite (1999) associated with considerable uncertainty.

For the sandstone, a value of 1 m^3 kg^{-1} was used previously. It does not seem appropriate to have higher sorption in the sandstone. Therefore, it is proposed that 0.5 m^3 kg^{-1} is appropriate. This new value takes into consideration a value of 0.35 m^3 kg^{-1} from Nirex (2003) and the value of 0.5 m^3 kg^{-1} within Quantisci (1999) for Zr, which may be considered to behave analogously to thorium.

4.11. Uranium

4.11.1. Geochemistry

Uranium aqueous chemistry is dominated by the IV and VI oxidation states. As indicated in Table 7 the distribution of uranium in a representative present day groundwater would be characterised by the predominance of U(VI), in the form of uranyl carbonates. Similarly, speciation calculations have been carried out under the conditions used in the site specific sorption experiments, with the result that the speciation is dominated by uranyl carbonates and hydroxides (Randall, 2002).

Redox conditions and pH would be expected to play a role in determining the sorption behaviour of uranium. Sharp sorption edges have been observed for many uranyl – mineral interactions, with maximum sorption commonly above pH ~6, e.g. gibbsite and silica (McKinley *et al*., 1995), smectite (Turner *et al*., 1996), ferrihydrite (Hsi and Langmuir, 1985; Waite *et al*., 1994) and quartz (Waite *et al*., 1993).

The presence of carbonate has been noted to have a profound effect on the sorption of uranium, with a marked decrease at pHs of neutral and above. This has been explained variously (e.g. Hsi and Langmuir, 1985; Waite *et al*., 1994; Payne and Waite, 1991;

Venkataramani and Gupta, 1991; Kohler *et al*., 1992) as being due to the formation of aqueous uranyl carbonate species, sorption of aqueous uranyl carbonates, formation of ternary surface uranyl carbonates or competition due to the sorption of carbonate itself).

The available site specific sorption data are quite extensive, particularly for sandy/coarse sediments. In addition, a reasonable range of pH values has been considered (6 – 8.5), which may allow some investigation of the effect of pH on sorption of uranium. However, as a result of the review presented in Randall *et al.* (2004b), a more limited set of experimental data are considered.

Figure 9 illustrates all the available sorption data for uranium onto LLWR sand, as a function of pH. It can be seen that there is a general decrease in K_d as pH increases, although there is a reasonable level of scatter. Of the three studies that explicitly looked at pH variation, Berry *et al.* (1990) and Haigh *et al*. (1989) both indicated lower sorption at high pH, whereas the behaviour in Yule (1994) depended on the presence of natural organics in the groundwater. These latter results indicate that in the presence of fulvic acid, the K_d of uranium decreases with increasing pH. However, the opposite is true for sorption when fulvic acid is not present.

The importance of uranium within the 2002 PCSC has meant that significant work has been undertaken, focussed on further understanding the behaviour of uranium in the geosphere environment. In particular, two sets of experimental studies have been undertaken to examine the sorption of uranium onto geosphere sediments. In the first of these, sorption onto a sample obtained from borehole 6125 (depth of 25.2m – mineralogical details of this sample are shown in Table 3 and Table 5) was examined as a function of pH and CO_2 partial pressure. Results are reported in Randall *et al*. (2004b).
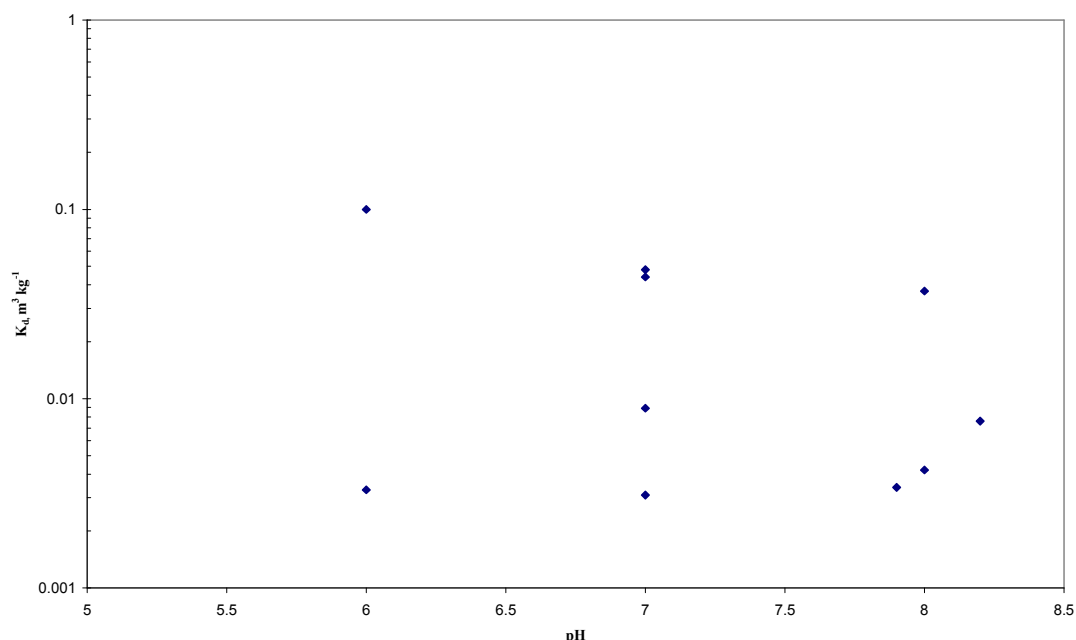
Figure 9: Uranium K_d as a function of pH from site specific sorption experiments onto coarse sediments.

More recent studies have been undertaken as part of the LLWR Lifetime Project (Dutton and Trivedi, 2008). Here, sorption was measured on two sediments, one clay-rich sediment and one sand. Mineralogical details are shown in Table 4.

Both of these studies indicate that the sorption of uranium is heavily dependent on pH, with sorption maxima around pH 6, and the presence of CO_2, with sorption generally lower under a CO_2 atmosphere of 1% (consistent with the conditions found in the LLWR geosphere).

These studies involved the use of well characterised sediments and so are ideally suited to the development of models to represent the sorption processes. Consequently, surface complexation models have been constructed to represent these reactions.

Two approaches have been utilised (see NEA, 2005). The first of these is the "component additivity" (or "bottom up") approach, where the surface mineralogy is characterised and parameters obtained from those of pure mineral phases. The second is the "generalised composite" (or "top down") model, where it is assumed that the composition of the surface mineralogy is inherently too complex to characterise and instead generic surface chemistry groups are used, with parameters obtained by fitting to experimental data.

The work reported in Randall *et al.* (2004b) utilised a component additivity approach, whereby a surface complexation model was constructed assuming that uranium sorption was dominated by ferrihydrite, an amorphous iron oxide, or goethite, a poorly crystalline phase. Both of these phases are considered to represent weathering products in the geosphere and have been identified during mineralogical characterisation. Thermodynamic constants to represent the sorption of uranium were taken from literature values for the pure mineral phases.

Example results are shown in Figure 10. This shows that the models provide a reasonable representation of the general features of uranium sorption, particularly the maximum at pH 6. However, the overall shape of the sorption curve does not reproduce the experimental results, which do not drop off to the same extent above pH 7.
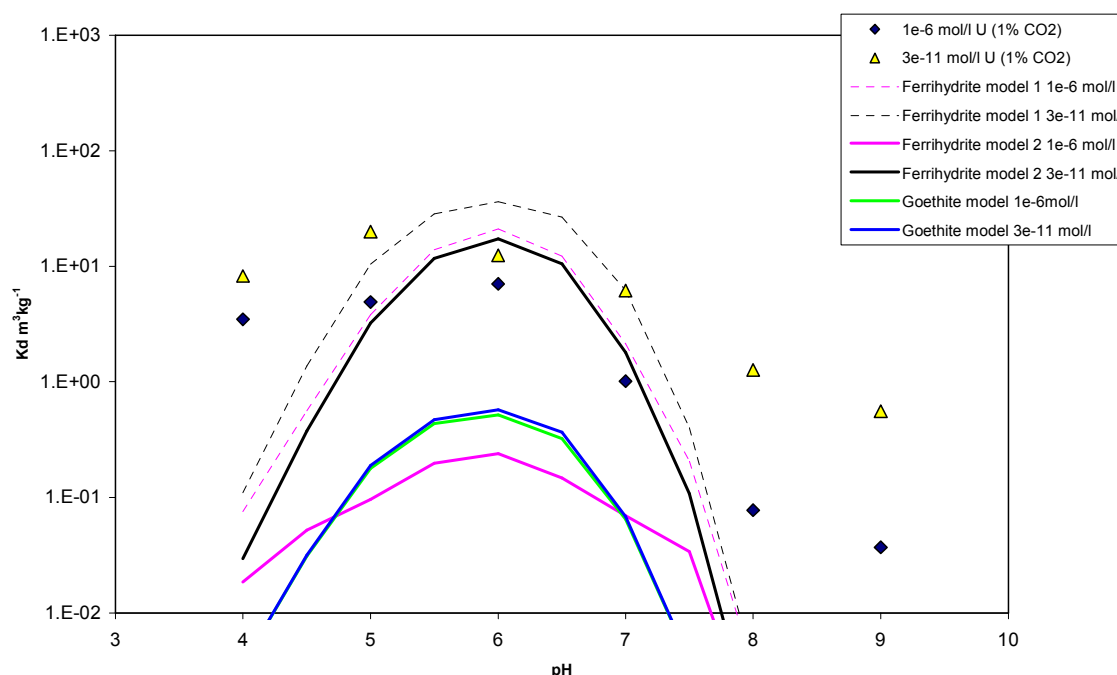
Figure 10: Comparison of experimentally measured sorption distribution coefficient (Kd) and modelled Kd for system with 1% CO2 (Randall et al., 2004b).

Consequently, in representing the experimental results of Dutton and Trivedi (2008), a more pragmatic, top down approach was taken. Here, no assumption was made of the exact nature of the uranium-mineral interactions. Instead the model was constructed to represent sorption of uranium onto "generic" surface sites, the concentrations of which were estimated from the surface areas of the sediments. Surface reactions were derived and fitted to reproduce the experimental data, with the guiding principle of chemically plausible surface species.

Of the number of models tested, two were found to fit the data well:

- Model 1 involved one uranium surface species: $SO\text{-}UO_2^+$, where S represents the surface.

- Model 2 involved two surface species: $SO\text{-}UO_2^+$ and $SO\text{-}UO_2CO_3^-$.

The results of this modelling exercise are shown in Figure 11 (for the case where CO_2 is absent) and Figure 12 (for the system with 1% CO_2). It can be seen that these relatively simple models reproduce the experimental data extremely effectively over most of the pH range.

To provide further confidence, the model fitted to the experimental results of Dutton and Trivedi (2008) was applied to the experimental results reported in Randall *et al.* (2004b). This is shown in Figure 13, where excellent agreement with experimental data is obtained.

It is extremely encouraging that a simple surface complexation model involving just two surface species is able to reproduce a wide range of experimental conditions. The two experimental studies represent extremes in the surface areas encountered within the LLWR geosphere (1.83 and 13.1 m^2 g^{-1}) and the success of the model means that it is

possible to interpolate between these sediments in order to estimate the K_d for a wide range of sediment types and pH ranges. For example, at pH 7.5 and a CO_2 partial pressure of 1%, the model predicts K_d values of 3.55 ml g^{-1} (or 0.00355 m^3 kg^{-1}) and 337 ml g^{-1} (or 0.337 m^3 kg^{-1}).
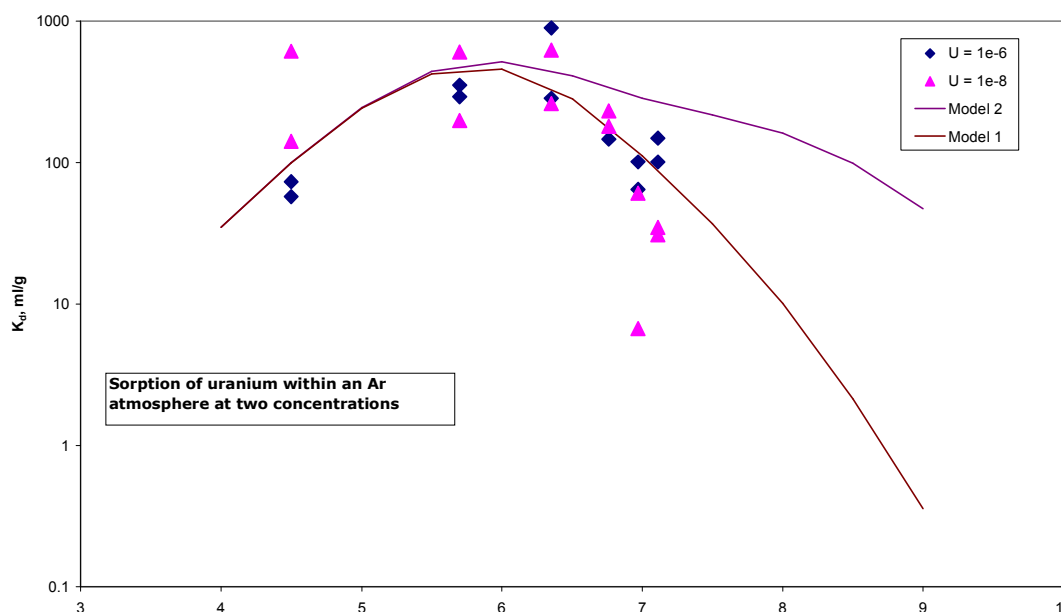
Figure 11: Comparison of modelled results from a simple surface complexation model and experimentally determined K_ds (no CO_2) (Dutton and Trivedi, 2008).
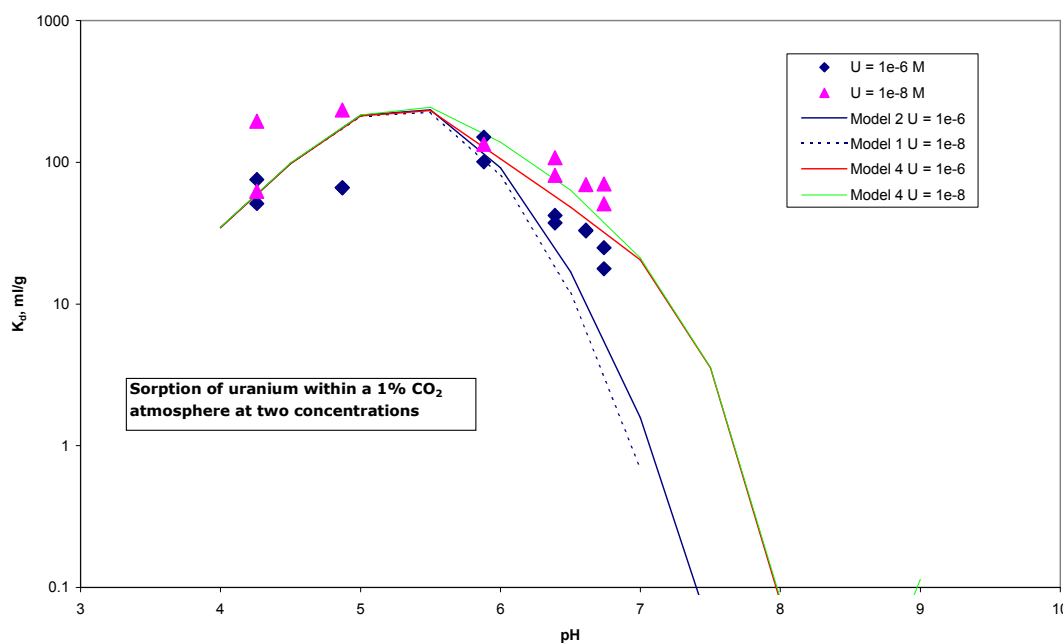
Figure 12: Comparison of modelled results from a simple surface complexation model and experimentally determined K_ds (1% CO_2) (Dutton and Trivedi, 2008).
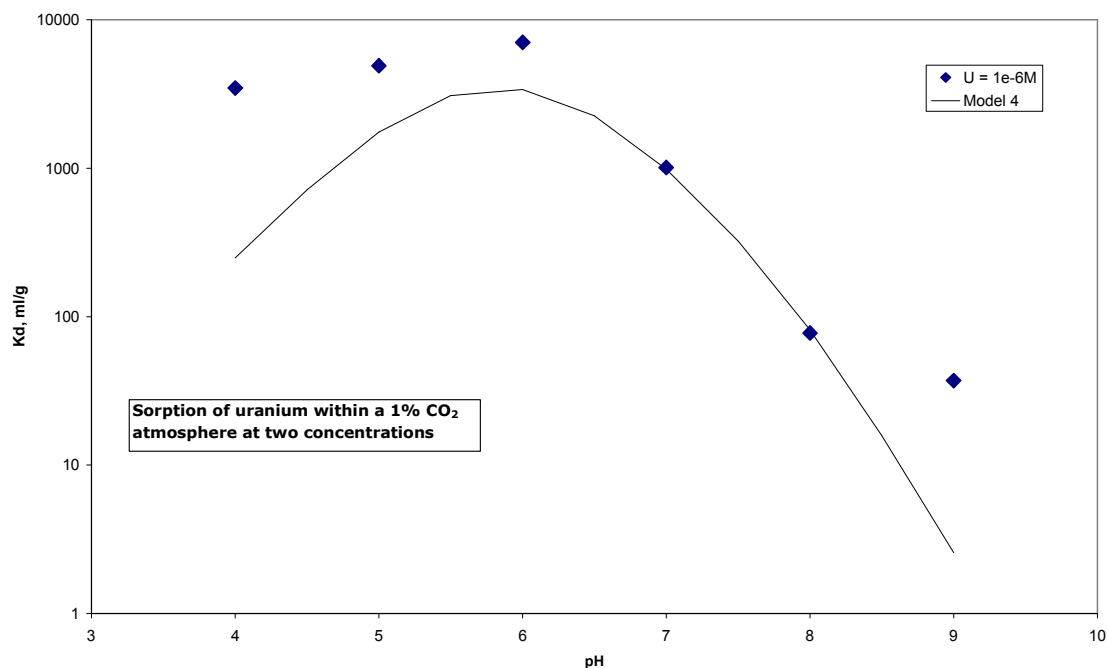
Figure 13: Comparison of modelled results from a simple surface complexation model and experimentally determined K_ds (1% CO_2) (Randall *et al.*, 2004b).

4.11.2.Recommended sorption parameters

The K_d used in the 2002 PCSC was determined from the geometric mean of a wide range of sorption data for coarse sediments, with a value of 0.16 m^3 kg^{-1}.

However, as a result of the review presented in Randall *et al.* (2004b), a more limited set of experimental data is considered. These are listed below in Table 8.

Of these, the determinations at pH 6 and pH 8 are not considered. Additionally, the Berry *et al.* (1990) data between "pH 7.5 to 7.9" are also not appropriate. One of these experimental values is the result of sorption onto "fines" separated from a sand sediment and so is not appropriate for representing sorption onto coarse sediments at the LLWR. The second measurement was taken at pH 7.9, which is considered to be too high. One of the Yule (1994) experiments did not report pH. Therefore, the most appropriate data are from Trivedi (1993) and two values from Yule (1994). The geometric mean for these data is 0.015 m^3 kg^{-1}. This fits well with the range of K_d predicted by the model presented in the previous section.

Only one site specific measurement of sorption onto silty sediments has been made (Braithwaite, 1999), giving a K_d of 0.03 m^3 kg^{-1}. However, this measurement could be complicated by possible precipitation of $CaUO_4$ and must therefore be viewed with caution.

For the sandstone, a slightly lower sorption value is deemed appropriate: 0.01 m^3 kg^{-1}, which is the same value as used previously in the 2002 PCSC (and a value 33% lower than that for drift sediments).

Table 8: Summary of site specific sorption data.

Sediment type	K_d (m³ kg⁻¹)				Comments	Reference
	pH 6	pH 7.0 to 7.4	pH 7.5 to 7.9	pH 8		
Coarse						
	0.093 (0.008)	0.048 (0.005)		0.037 (0.004)		Yule 1994
	0.104 (0.010)	0.043 (0.005)		0.004 (0.002)	+10 ppm fulvic acid	Yule 1994
		? 0.162 (0.013)			pH not specified	Yule 1994
		0.0089 (0.0018)			Initial HCO₃:1.39E-03 M	Trivedi 1993
		0.0031 (0.0001)			Initial HCO₃: 0.016 M	Trivedi 1993
				0.0076 (0.0010)		Berry et al., 1990
			0.0034 (0.0002)		+ Fluka humic acid	Berry et al., 1990
			6.8 (0.6)		Used 'fines' at M/V 1:100	Berry et al., 1990

4.12. Neptunium

4.12.1. Geochemistry

The chemistry of neptunium is dependent on redox conditions and, as indicated in Table 7, neptunium is predicted to exist mostly in the Np(V) oxidation state with some contribution from the Np(IV) oxidation state. Speciation is dominated by the NpO_2^+ species. All of the site specific sorption experiments were carried out under aerobic conditions, with speciation dominated by Np(V) species, such as NpO_2^+ and $NpO_2CO_3^-$ (Randall, 2002).

Under oxic conditions the sorption of neptunium onto single minerals behaves similarly to many metal – mineral interaction systems, in that a sharp sorption edge is observed (e.g. Girvin et al., 1991; Nakayama and Sakamoto, 1991; Kohler et al., 1999). The effect of carbonate appears to be less than that observed for uranium, for example. A simple surface complexation model has been used quite successfully by Bradbury and Baeyens (1992; 1993) to model the sorption of neptunium onto a range of natural mineral phases.

Insufficient data are available to indicate controlling factors over sorption behaviour of neptunium, although Haigh et al. (1989) conclude that complexation with natural organics is unlikely to occur.

4.12.2. Recommended sorption parameters

For the 2002 PCSC, the results of Stafford (1995), Trivedi (1993) and Haigh *et al.* (1989) were used in deriving a K_d for coarse sediments. However, given that there are questions regarding the experimental methodology of Stafford (1995), and the Haigh *et al.* measurements were taken at pH 8.3, only the measured K_ds from Trivedi (1993) are deemed appropriate. Two values were measured (0.02 and 0.01 m^3 kg^{-1}) giving a geometric mean of 0.014 m^3 kg^{-1}.

Only one site specific datum point is available for silt sediments (Braithwaite, 1999), with a K_d of 0.032 m^3 kg^{-1}.

For the sandstone, a value of 0.005 m^3 kg^{-1} was used in the 2002 PCSC. However, the neptunium K_d for drift sediments is similar to that of uranium. Therefore, rather than using the value of 0.005 m^3 kg^{-1}, it is recommended that the uranium sandstone K_d of 0.01 m^3 kg^{-1} is used for neptunium in the radiological assessment.

4.13. Plutonium

4.13.1. Geochemistry

Plutonium can exist in a number of oxidation states, however, speciation calculations show that Pu(IV) is the dominant species in the LLWR reference groundwater (Table 7), with Pu(CO$_3$)$_3^{2-}$ predicted to dominate. Thus, the impact of redox conditions will not directly affect the sorption of plutonium.

Variations in pH are likely to be more significant in terms of determining plutonium sorption. For example, Sanchez *et al.* (1985) showed a strong pH dependence for the sorption of Pu(IV) onto goethite, with a sorption edge observed between pH 3-5. These workers also demonstrated that the presence of carbonate has an influence on plutonium sorption, with sorption decreasing above a total alkalinity of 100 meq l^{-1}. This was attributed to the formation of plutonium carbonate aqueous species.

Figure 14 shows the measured K_d values as a function of pH, for site specific coarse sediments. This shows a minimal trend in sorption when all the data are taken into account, although there is some evidence of slight variation from the experiments of Yule (1994). Additionally from the Yule (1994) experiments, the presence of fulvic acid increases the K_d at all pHs (in contrast to the corresponding uranium results).

From Trivedi (1993), sorption onto "G4 sand" is two orders of magnitude greater than that observed for "G6 sand". This may be due to the increased carbonate concentration in the G6 experiment.
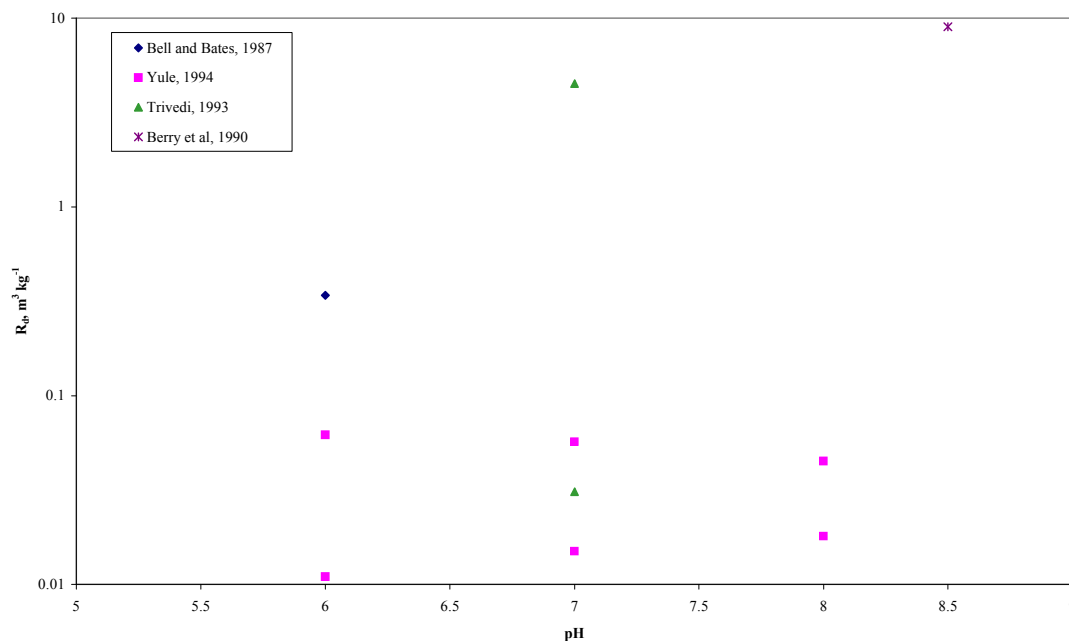
Figure 14: Plutonium sorption onto LLWR sand as a function of pH.

4.13.2.Recommended sorption parameters

For the 2002 PCSC, the results of Stafford (1995), Trivedi (1993), Yule (1994), Bell and Bates (1987) and Berry *et al.* (1990) were used in deriving a K_d for coarse sediments. However, there are questions regarding the experimental methodology of Stafford (1995) and Berry *et al*. (1990), and some of the Yule, Bell and Bates measurements were taken at inappropriate pHs. Therefore, only the measured K_ds from Trivedi (1993) and the measurements of Yule at pH 7 are deemed appropriate. Two values were measured (4.5, 0.031, 0.015 and 0.057 m^3 kg^{-1}) giving a geometric mean of 0.10 m^3 kg^{-1}.

Site specific data comes from just one source (Braithwaite, 1999), with a measured K_d of 6.6 m^3 kg^{-1}.

For the sandstone, a value of 0.1 m^3 kg^{-1} was used previously. Given the reduction assumed for other actinides, a K_d of 0.07 m^3 kg^{-1} is deemed appropriate.

4.14. Americium

4.14.1.Geochemistry

Americium exists in one oxidation state in aqueous conditions, and so any changes in redox conditions will have little impact. In terms of pH variation, Stammose and Dolo (1990) examined the sorption of americium onto clay minerals, with the result that K_d increased steadily between pH 2-6, and remained constant thereafter. Allard *et al.* (1989) noted that the presence of fulvic and humic material decreased the sorption of americium onto alumina.

Very little data are available to examine possible mechanisms and controlling factors for site specific material. In contrast to the Allard *et al.* study, Stafford (1995) found that sorption onto LLWR sand was little affected by the presence of fulvic acid, with the presence of fulvic acid reducing the K_d from 0.15 to 0.08 m^3 kg^{-1}.

4.14.2. Recommended sorption parameters

For the 2002 PCSC, only the results of Stafford (1995) and Trivedi (1993) were used in deriving a K_d. However, given that there are questions regarding the experimental methodology of Stafford (1995), only the measured K_ds from Trivedi are deemed appropriate. Two values were measured (0.17 and 0.11 m^3 kg^{-1}) giving a geometric mean of 0.14 m^3 kg^{-1}.

Only one site specific source of data is available for silt sediments (Braithwaite, 1999) with a K_d of 46 m^3 kg^{-1}. However, the experimental error associated with this measurement is very large (± 66 m^3 kg^{-1}), thus it is difficult to recommend use of this value.

For the sandstone, a value of 0.1 m^3 kg^{-1} was used in the 2002 PCSC. This seems a reasonable value to apply to the sandstone, which represents a value approximately 30% lower than that for the drift sediments.

4.15. Analogues

There are no site specific sorption data for a number of radionuclides of interest to the current radiological assessment. Consistent with the approach used in the 2002 PCSC, it has been decided to use data from analogue elements rather than rely on generic data.

The following table (Table 9) lists these elements and provides justifications for the use of particular analogues.

Table 9: Summary of analogues used for radionuclides with no site specific data.

Radionuclide	Analogy to	Assumed Zero	Justification
Cl		✔	Chlorine exists as Cl^- in the aqueous environment and exhibits little tendency to form complexes, or undergo sorption.
Cm	Am		Like americium, Cm predominantly exists in the 3+ oxidation state, has a similar electronic structure (Am = [Rn] $5f^7$ $7s^2$, Cm = [Rn] $5f^7$ $6d^1$ $7s^2$), and almost identical ionic radii, in the 3+ oxidation state (Am = 97.5 pm, Cm = 97 pm).
Pa	Pu		Pa has a high first hydrolysis constant and a high z/d ratio and so should also exhibit strong sorption at high pH. The chemistry of Pa, in terms of the formation of 4+ and 5+ oxidation states, is similar to Np or Pu. From generic sorption databases (e.g. Shepperd and Thiabault, 1990; Quantisci, 1999; Nirex, 2003) sorption behaviour is closer to that of Pu.
Ac	Am		As actinium exists predominantly in the 3+ oxidation state, it is expected to behave in a similar manner to americium.
Nb	Zr/Th		Niobium is expected to exhibit similar behaviour to zirconium, being also a second row transition metal. Speciation calculations indicate the predominance of the $Nb(OH)_5^0$ species, which, as a neutral hydroxide species, is similar to the speciation of Zr.
Pb	Ni		The aqueous chemistry of lead is likely to be dominated by the Pb(II) oxidation state. It forms complexes with a variety of inorganic ligands, such as carbonate and hydroxyl species. Therefore it is proposed to use nickel data as an analogue.
Zr	Th		The domination of $Zr(OH)_4$ over the expected pH range for the LLWR geosphere (Randall, 2002) would indicate that thorium is a suitable analogue.

5.   Summary

The previous sections have described the derivation process for assigning K_ds for each radionuclide of interest in the radiological assessment of the groundwater pathway for the LLWR (Paksy and Henderson, 2008), representing an update on the 2002 PCSC data. Where data were not available, analogues have generally been used.

Table 10 shows a summary of all of these K_ds for both the drift sediments and the sandstone.

Table 10: Updated sorption coefficients (m^3 kg^{-1}) for the LLWR geosphere.

Species	Drift	Sandstone
Ac	1.4E-01	1.0E-01
Am	1.4E-01	1.0E-01
C	5.0E-04	5.0E-04
Cl	0.0E+00	0.0E+00
Cm	1.4E-01	1.0E-01
Co	1.4E-01	1.0E-01
Cs	3.0E-01	3.0E-01
H	0.0E+00	0.0E+00
I	1.1E-03	0.0E+00
Mo	1.0E-03	1.0E-04
Nb	8.8E-01	5.0E-01
Ni	4.0E-02	3.0E-02
Np	1.4E-02	1.0E-02
Pa	1.0E-01	7.0E-02
Pb	4.0E-02	1.0E-02
Pu	1.0E-01	7.0E-02
Ra	8.6E-02	5.0E-02
Sr	1.2E-02	7.5E-03
Tc	1.0E-04	1.0E-04
Th	8.8E-01	5.0E-01
U	1.5E-02	1.0E-02
Zr	8.8E-01	5.0E-01

References

Akhurst, M.C., Chadwick, R.A., Holliday, D.W., McCormac, M., McMillian, A.A., Millward, D., Young, B., Ambrose, K., Auton, C.A., Barclay, W.J., Barnes, R.P., Beddoe-Stephens, B., James, J.W.C., Johnson, H., Jones, N.S., Glover, B.W., Hawkins, M.P., Kimbell, G.S., MacPherson, K.A.T, Merritt, J.W., Milodowski, A.E., Riley, N.J., Robins, N.S. Stone, P. and Wingfield, R.T.R. 1997. The geology of the west Cumbria district. Memoir of the British Geological Survey. Sheets 28, 37 and 47 (England and Wales).

Appelo, C. A. J and Postma, D., 1993. Geochemistry, groundwater and pollution. A.A. Balkema, Rotterdam, Netherlands.

Allard, B., Moilin, V., Basso, L., Tran, M. T. and Stammose, D., 1989. Americium adsorption onto alumina in the presence of humic materials. *Geoderma,* **44**, 181 – 187.

Bell, J. and Bates, T. H., 1987. Distribution coefficients of radionuclides between soils and groundwaters and their dependence on various test parameters. BNFL Report. Chemistry Group Memorandum 160.

Berry, J. A., Brownsword, M., Gilling, D., Jefferies, N. L., Lineham, T. R. and Linsell, I. M., 1990. Mass transfer and sorptive properties of geological samples from the Drigg site. AEA Technology Report AERE R 13131.

Bloodworth, A. J. and Inglethorpe, S. D. J., 1988. Mineralogy of shallow borehole material from Drigg, Cumbria. British Geological Survey Mineralogy and Petrology Report WG/88/5C.

BNFL, 2002a. Drigg Operational Environmental Safety Case.

BNFL, 2002b. Drigg Post-Closure Safety Case: Overview Report.

BNFL, 2002c. Drigg Post-Closure Safety Case. Geological Interpretation

BNFL, 2002d. Drigg Post-Closure Safety Case: Hydrogeological Interpretation.

BNFL, 2002e. Drigg Post-Closure Safety Case: Geochemical Interpretation.

Bradbury, M. H. and Baeyens, B., 1992. A mechanistic approach to the generation of sorption databases. In: "Radionuclide Sorption from a Safety Assessment Perspective" Proceedings of a Nuclear Energy Agency Workshop, OECD, Paris, pp121 – 162.

Bradbury, M. H. and Baeyens, B., 1993. A general application of surface complexation to modeling radionuclide sorption in natural systems. *J. Colloid and Interface Science* **158** 364-371.

Bradbury, M. H. and Baeyens, B., 2000. A generalised sorption model for the concentration dependent uptake of caesium by argillaceous rocks. *J. Contaminant Hydrology*, **42**, 141-163.

Braithwaite, A., 1997. The Sorption Behaviour of U, Sr, Cs, Pu and Am on a clay-rich sediment in the presence of sodium perchlorate. BNFL Internal Report RDR 1156.

Braithwaite, A., 1999. The sorption behaviour of radionuclides on a silty sediment. BNFL Internal Report RDR 1377.

de Preter, P. D., Loon, L. V., Maes, A. and Cremers, A., 1991. Solid liquid distribution of radiocesium in Boom Clay; A quantitative interpretation. *Radiochimica Acta*, **52/53**, 299-302.

Dutton, M. and Trivedi, T. 2008. Sorption of uranium onto geosphere sediments. Nexia Solutions Report (08) 9367.

Eilbeck, A., 1999. Some distribution ratio data for the sorption of caesium and strontium on Drigg clay. DTP Technical Note TN59.

Environment Agency, Scottish Environmental Protection agency and the Department of the Environment for Northern Ireland, 1997. Radioactive Substances Act 1993 – Disposal facilities on land for low and intermediate level radioactive wastes: Guidance on requirements for authorisation. Bristol, Environment Agency.

Girvin, D. C., Ames, L. L., Scwab, A. P. and McGarrah, J. E., 1991. Neptunium adsorption on synthetic amorphous iron okyhydroxides. *J. Colloid and Interface Science*, **141**, 67-78.

Grütter, A., von Gunten, H. R., Rössler, E. and Keil, R., 1994. Sorption of Strontium on Unconsolidated Galaciofluvial Deposits and Clay Minerals: Mutual Interference of Cesium, Strontium and Barium. *Radiochimica Acta*, **64**, 247-252.

Haigh, D. G. and Hooker, P. J., 1988. A Sorption Study of Sr-85, Cs-137 and Th-227 onto glacial sand as part of an inter - laboratory exercise. DoE Report DOE/RW/88/078.

Haigh, D. G., Williams, G. M., Hooker, P. J., Ross, C. A. M., Allen, M. R. and Warwick, P., 1988. The effect of organics on the sorption of cobalt by glacial sand in laboratory batch experiments. British Geological Survey Technical Report WE/88/20/.

Haigh, D. G., Higgo, J. J. W., Williams, G. M., Hooker, P. J., Ross, C. A. M., Falck, W. E. and Allen, M. R., 1989. The effect of organics on the sorption of Strontium, Caesium, Iodine, Neptunium, Uranium and Neptunium by glacial sand. DoE Report DOE/RW/89.067.

Henrion P.N., Monsecour M., Fonteyne A., Put M. and Deregge P., 1985. Migration of radionuclides in Boom Clay. *Radioactive Waste Management and The Nuclear Fuel Cycle*, **8(2)**, 143 - 159.

Hsi, C-K. D., and Langmuir, D., 1985. Adsorption of uranyl onto ferric oxyhydroxides: Application of the surface complexation site-binding model. *Geochimica et Cosmochimica Acta*, **49**, 1931-41.

Hunter, J., Michie, U.Mcl., Smith, N.T. and Towler, G., 2008. LLWR Lifetime Project: Reinterpretation of the Quaternary geology of the LLWR Site and the surrounding region. Nexia Solutions Report No. 8794.

Kohler, M., Wieland, E. and Leckie, J. O., 1992. Metal-ligand surface interactions during sorption of uranyl and neptunyl on oxides and silicates. In: Water-Rock Interaction. Eds. Kharaka and Maest, Balkema, Rotterdam.

Kohler, M., Honeyman, B. D., and Leckie, J. O. 1999. Neptunium (V) sorption on hematite. *Radiochimica Acta* **85**, 33.

LaFlamme, B. D. and Murray, J. W., 1987. Solid/Solution interaction: the effect of carbonate alkalinity on adsorbed thorium. *Geochimica et Cosmochimica Acta*, **51**, 243 – 250.

Loftus, C., 1995. Mineralogical characterisation of selected glacial drift deposits. MSc Thesis, Department of Earth Sciences, University of Manchester.

Merritt, J. W. and Auton, C.A., 2000. An outline of the lithostratigraphy and depositional history of Quaternary deposits in the Sellafield district, west Cumbria. Proceedings of the Yorkshire Geological Society, Vol. 53, pp 129-154.

McKinley, I. G., and West, J., 1981. Radionuclide sorption/desorption processes occurring during groundwater transport - Progress report Oct. 1979 - Dec. 1980. BGS Report No. ENPU 81-6.

McKinley, J. P., Zachara, J. M., Smith, S. C. and Turner, G. D., 1995. The influence of uranyl hydrolysis and multiple site-binding reactions on adsorption of U(VI) to montmorillonite. *Clays and Clay Minerals*, **43**, 586 – 598.

Milodowski, A. E., 1990. Petrographic and sedimentological characteristics of drift sediments from the radiotracer experiment array at Drigg, Cumbria. British Geological Survey Technical Report WE/90/24.

NAGRA, 2002. NAGRA Project Opalinus Clay. Safety Report. Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste (Entsorgungsnachweis). Nagra Technical Report 02-05. 2002.

Nakayama, S. and Sakamoto, Y., 1991. Sorption of neptunium on naturally-occurring iron-containing minerals. *Radiochimica Acta*, **52/53**, 153-157.

NEA, 2005. NEA Sorption Project Phase II: Interpretation and prediction of radionuclide sorption onto substrates relevant to radioactive waste disposal using thermodynamic sorption models. Nuclear Energy Agency/OECD, Paris.

Nirex 2003. Generic repository studies. Generic post-closure performance assessment. Nirex Report N/080.

Payne, T. E. and Waite, T. D., 1991. Surface complexation modelling of uranium sorption data obtained by isotope exchange techniques. *Radiochimica Acta*, **52/53**, 487-493.

Plummer, M. A, Hull, L. C, and Fox, D. T. 2004. Transport of Carbon-14 in a Large Unsaturated Soil Column. Vadose Zone Journal 3:109–121.

Polfer, N. and Braithwaite, A., 1998. Nickel transport through sand columns. Results presented in Braithwaite and Prastka (1998b).

Paksy, A. and Henderson, E. 2008. LLWR Lifetime Project: Assessment of radiological impacts for the groundwater pathway. Nexia Solutions Report 9449.

Prastka, K., 1999. An experimental and literature review of some aspects of the geochemistry of Drigg. DTP Technical Note 041.

Quantisci, 1999. Assessment of the consequences of the presence of toxic elements in some common radioactive waste streams. Report produced for the European Commission Ref. EUR 18211.

Rancon, D., 1988. Comparative study of radioactive iodine behaviour in soils under various experimental and natural conditions. *Radiochimica Acta,* **44/45,** 187-193.

Randall, M.G., Trivedi, D.P., Graham, J. Small, J.S and Hughes C. 2004a. Mineralogical characterisation of sediments at the Drigg low level radioactive waste disposal site and the influence on groundwater chemistry. *Materials Research Society Symposium, Scientific Basis for Nuclear Waste Management XXVII* **807,** p557-562.

Randall, M. G., Small, J. S., and Sanchez, A. 2004b. Further investigations of uranium sorption in the Drigg geosphere. NSTS Report 5187.

Randall, M., 2002. Site specific sorption database for the Drigg geosphere. Drigg Technical Programme DTP 02/03 TN 040.

Sanchez, A. L., Murray, J. W. and Sibley, T. H., 1985. The adsorption of plutonium IV and V on goethite. *Geochimica et Cosmochimica Acta*, **49**, 2297 – 2307.

Sawhney, B. L., 1972. Selective sorption and fixation of cations by clay minerals – a review. *Clay Minerals* **20** p93.

Sheppard, M. I. and Thiabault, D. H., 1990. Default Soil Solid/Liquid Partition Coefficients, K_d's for Four Major Soil Types: A Compendium. *Health Physics*, **59(4)**, 471.

Smith, N.T., 2008. LLWR Lifetime Project: Phase II Geological Reinterpretation: Further 3D geological modelling of the Quaternary geology of the LLWR Site and the surrounding region. Nexia Solutions Report No. (07)8885 Issue 2.

SKB. 2004. Interim data report for the safety assessment SR-Can. SKB Report R-04-34.

Smith, N.T., 2008. LLWR Lifetime Project: Phase II Geological Reinterpretation: Further 3D geological modelling of the Quaternary geology of the LLWR Site and the surrounding region. Nexia Solutions Report No. (07)8885 Issue 2.

Stafford, C. J., 1993. Uranium sorption onto G6 sand. BNFL internal report. RDR 349.

Stafford, C. J., 1995. Radionuclide sorption onto G6 sand. BNFL Internal Report RDR 619.

Stammose, D. and Dolo, J.-M., 1990. Sorption of americium at trace level on a clay mineral. *Radiochimica Acta*, **51**, 189-193.

Stenhouse, M. J., 1995. Sorption Databases for Crystalline, Marl and Bentonite for Performance Assessment. Nagra Technical Report 93-06.

Strickert, R. G., Friedman, A. M. and Fried, S., 1980. The sorption of technetium and iodine radioisotopes by various minerals. *Nuclear Technology*, **49**, 263-266.

Thompson, G. and Miller, B. 2005. SFR 1 – Post closure radionuclide release and dose calculations. SKB Report. R-05-81.

Torstenfelt, B., Andersson, K. and Allard, B., 1982. Sorption of Strontium and Cesium on Rocks and Minerals. *Chemical Geology*, **36**, 123-137.

Trivedi, D. P., 1993. Batch Sorption of Pu, U, Am, Sr and Np on Drigg far field sediments. BNFL TD Memo 332.

Turner, G. D., Zachara, J. M., McKinley, J. P. and Smith, S. C., 1996. Surface-charge properties and UO_2^{2+} adsorption of a subsurface smectite. *Geochimica et Cosmochimica Acta*, **18**, 3399 – 3414.

USGS, 2001. Surface complexation modelling of uranium(VI) adsorption on natural mineral assemblages. NUREG/CR-6708. U.S. Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, DC, 20555-0001.

Venkataramani, B. and Gupta, A. R., 1991. Effect of anions on the sorption of uranyl ions by hydrous oxides: Application of the surface hydrolysis model. *Colloids and Surfaces*, **53**, 1-19.

Waite, T. D., Payne, T. E., Davis, J. A. and Sekine, K. 1993. Uranium sorption modelling – A surface complexation approach. 5th Commission of the European Communities

Natural Analogue Working Group Meeting and Alligator River Analogue Project (ARAP) Final Workshop. Ed. H. von Maravic and J. Smellie, Brussels, Belgium.

Waite, T. D., Davis, J. A., Payne, T. E., Waychunas, G. A. and Xu, N., 1994 Uranium (VI) adsorption to ferrihydrite: Application of a surface complexation model. *Geochimica et Cosmochimica Acta*, **58**, 5465-5478.

Williams, G. M., Alexander, L. S., Hitchman, S. P., Hooker, P. J., Noy, D. J., Ross, C. A. M., Stuart, A. and West, J. M., 1985. In-situ radionuclide migration studies in a shallow sand aquifer. British Geological Survey Report FLPU 85-7.

Yule, L., 1994*.* Batch sorption experiments: Cs, U, Pu, Tc, Cu and Ni onto G6 sand. BNFL internal report. TDM 625.

Appendix A

Table A1: Data sources for site specific sorption

Radionuclide	Sediment	Data source
Cs	Coarse	McKinley & West (1981) Bell & Bates (1987) Haigh & Hooker (1988) Haigh *et al.* (1989) Yule (1994)
	Silt	Haigh & Hooker (1988) Braithwaite (1999)
	Clay	Bell & Bates (1987) Braithwaite (1997) Eilbeck (1999)
Sr	Coarse	Bell & Bates (1987) Haigh & Hooker (1988) Haigh *et al.* (1989) Berry *et al.* (1990) Trivedi (1993) Eilbeck (1999)
	Silt	Haigh & Hooker (1988) Braithwaite (1999)
	Clay	Braithwaite (1997) Eilbeck (1999)
Ra	Coarse	Stafford (1995)
	Silt	Braithwaite (1999)
	Clay	None
Co	Coarse	McKinley and West (1981) Haigh *et al.* (1988)
	Silt	None
	Clay	None
Ni	Coarse	Yule (1994) Polfer and Braithwaite (1998)
	Silt	None
	Clay	None
Tc	Coarse	Yule (1994) Williams *et al.* (1985)
	Silt	None
	Clay	None

Radionuclide	Sediment	Data source
I	Coarse	Williams *et al.* (1985) Haigh *et al.* (1989
	Silt	None
	Clay	None
Th	Coarse	Bell and Bates (1987) Haigh and Hooker (1988) Stafford (1995)
	Silt	Haigh and Hooker (1988) Braithwaite (1999)
	Clay	Bell and Bates (1987)
U	Coarse	Bell and Bates (1987) Haigh *et al.* (1989) Berry *et al.* (1990) Trivedi, 1993) Stafford (1995) Yule (1994)
	Silt	Braithwaite (1999)
	Clay	Bell and Bates (1987) Berry *et al.* (1990) Braithwaite (1997)
Np	Coarse	Haigh *et al.* (1989) Stafford (1995) Trivedi (1993)
	Silt	Braithwaite (1999)
	Clay	None
Pu	Coarse	Haigh *et al,* (1989) Berry *et al.* (1990) Trivedi (1993) Yule (1994) Stafford (1995)
	Silt	Braithwaite (1999)
	Clay	Braithwaite (1997) Berry *et al.* (1990)
Am	Coarse	Stafford (1995) Trivedi (1993)
	Silt	Braithwaite (1999)
	Clay	Braithwaite (1997)

Table A2: Summary of groundwater compositions used in site specific sorption experiments

Groundwater	Reference	Ca^{2+}(M)	Mg^{2+}(M)	Na^+(M)	K^+(M)	Cl^- (M)	TIC (M)	SO_4^{2-} (M)	NO_3^- (M)	Fe^{3+}(M)	Ionic Strength (M)	pH	Eh (mV)
Groundwater from BHP8 (Sellafield)	Bell & Bates (1987)	9.48×10^{-4}	3.70×10^{-4}	1.1×10^{-3}	2.56×10^{-5}	1.52×10^{-3}	1.30×10^{-3}	3.12×10^{-4}	1.94×10^{-4}	-	~0.0073	5.7	+200
Drigg groundwater (DGW-B)	Haigh $et\ al.$ (1989)	1.2×10^{-3}	4.6×10^{-4}	9.7×10^{-4}	3.8×10^{-5}	9.6×10^{-4}	3.0×10^{-3}	2.5×10^{-4}	2.1×10^{-6}	$<1.8 \times 10^{-7}$	~0.0063	7.3	+400
Drigg groundwater from BH42	Berry $et\ al.$ (1990a)	2.9×10^{-3}	7.8×10^{-4}	1.5×10^{-3}	8.4×10^{-5}	3.8×10^{-3}	4.0×10^{-3}	3.1×10^{-4}	1.9×10^{-6}	$<7.1 \times 10^{-8}$	~0.0187	-	Aerobic
Drigg G6 water	Trivedi (1993)	5.24×10^{-4}	1.69×10^{-4}	0.017	$<6.39 \times 10^{-6}$	1.35×10^{-3}	0.016	1.25×10^{-4}	$<1.61 \times 10^{-6}$	2.33×10^{-5}	~0.0189	Adjusted to 7.0	Aerobic
Drigg G4 water	Trivedi (1993)	1.2×10^{-3}	8.6×10^{-5}	3.0×10^{-4}	$<1.3 \times 10^{-5}$	3.1×10^{-4}	1.39×10^{-3}	1.4×10^{-4}	1.9×10^{-5}	$<1.8 \times 10^{-5}$	~0.0039	Adjusted to 7.0	Aerobic
Simulated groundwater	Yule (1994)	4.43×10^{-4}	1.87×10^{-4}	5.52×10^{-4}	-	6.0×10^{-4}	1.08×10^{-3}	1.12×10^{-4}	1.60×10^{-6}	-	~0.0026	7.5	+140
Simulated Groundwater at end of experiment	Yule (1994)	4.24×10^{-3} (Ni) 8.98×10^{-4}(U) 8.23×10^{-4}(Cs)	1.83×10^{-4}(Ni) 2.18×10^{-4}(U) 2.18×10^{-4}(Cs)	1.2826×10^{-2}(Ni) 1.39×10^{-2}(U) 1.30×10^{-2}(Cs)	-	9.59×10^{-4}(Ni) 8.32×10^{-4}(U) 8.32×10^{-4}(Cs)	7.16×10^{-4}(Ni) 9.33×10^{-4}(U) 1.83×10^{-3}(Cs)	1.87×10^{-4}(Ni) 3.12×10^{-4}(U) 3.12×10^{-4}(Cs)	-	0 (Ni) 1.8×10^{-5}(U) 4×10^{-6}(Cs)	-	6,7,8	+140
Simulated groundwater + humics	Yule (1994)	1.12×10^{-3} (Pu,Tc) 3.36×10^{-4}(Ni) 3.5×10^{-4}(U,Cs)	3.68×10^{-4}(Pu,Tc) 1.89×10^{-4}(Ni) 1.93×10^{-4}(U,Cs)	1.06×10^{-2}(Pu,Tc) 1.30×10^{-2}(Ni) 1.30×10^{-2}(U,Cs)	-	1.39×10^{-3}(Pu,Tc) 8.18×10^{-4}(Ni) 9.66×10^{-4}(U,Cs)	2.61×10^{-3} (Pu,Tc) 1.22×10^{-3}(Ni) 8.74×10^{-4}(U,Cs)	3.9×10^{-4}(Pu,Tc) 2.19×10^{-4}(Ni) 2.19×10^{-4} (U,Cs)	-	0 (Pu,Tc) 4×10^{-6}(Ni) 2×10^{-6} (U,Cs)	-	6,7,8	+140
Simulated groundwater	Stafford (1995)	4.43×10^{-4}	1.87×10^{-4}	1.08×10^{-3}	-	8.86×10^{-4}	1.08×10^{-3}	1.87×10^{-4}	1.60×10^{-6}	-	0.0132	7.64(U) 7.50 (Pu) 6.73(Th) 7.63(Am) 7.52(Np) 7.70(Ra)	+550(U) -(Pu) +550(Th) +590(Am) +590(Np) +520(Ra)
Simulated Groundwater at end of experiment	Stafford (1995)	4.49×10^{-4}	2.06×10^{-4}	1.17×10^{-2}	1.05×10^{-4}	Not analysed	6.67×10^{-4}	2.49×10^{-4}	Not analysed	-	-	7.64 (U) 7.47 (Pu) 6.73(Th) 7.14(Am) 7.14(Np) 7.18(Ra	+550 (U) +500(Pu) +470(Th) +400(Am) +420(Np) +460(Ra)
Drigg groundwater	Haigh and Hooker (1988)	34.6 ppm	6.1 ppm	39.7 ppm	5.9 ppm	61.5 ppm	132 ppm	36.6 ppm	0.34 ppm	0.01 ppm		6	8
Simulated groundwater	Eilbeck (1999)	17.1 ppm	5.3 ppm	224 ppm	-	35.8 ppm	64.8 ppm	17.8 ppm	0.8 ppm	-	-	7.2	6.8
Drigg Simulated Groundwater	Braithwaite (1997)	7.21×10^{-4}	2.6×10^{-4}	1.76×10^{-3}	-	$9.13\ 10^{-4}$	$1.11\ 10^{-3}$	$2.04\ 10^{-4}$	-	-	-	7.4 (Pu) 7.4 (U) 7.6 (Cs, Sr) 6.9 (Am)	9.4 (Pu) 7.5 (U) 7.6(Cs, Sr) 7.7 (Am)
Drigg Simulated groundwater + 0.01M NaClO₄	Braithwaite (1997)	$6.37\ 10^{-4}$	$2.22\ 10^{-4}$	0.0118	-	$8.85\ 10^{-4}$	$1.19\ 10^{-3}$	$2\ 10^{-4}$	-	-	-	7.5(Pu) 7.3 (U) 7.6(Cs,Sr) 7 (Am)	9 (Pu) 7.6 (U) 7.8(Cs,Sr) 10.2 (Am)

Table A3: Experimental details for site specific sorption experiments.

Author	Experimental Details	M/V	Contact Time (d)	Vessel Type	Phase Separation
Braithwaite 1999	Batch - Duplicate samples continuously rotated at <10rpm	1:50	75	Polypropylene	Centrifugation (15mins) (3000rpm) Ultrafiltration (Amicon 30000 MWCO paper)
Stafford 1995	Batch - Duplicate samples continuously rotated at <10rpm	1:50	65	Polypropylene	Centrifugation (15mins) (3000rpm) Ultrafiltration (Amicon 30000 MWCO paper)
Trivedi 1993	Batch - Duplicate samples	1:5	10 - 14	DNA	Centrifugation, Filtration (0.02 micron filter)
Braithwaite 1997a	Batch - Duplicate samples continuously rotated at <10rpm	1:50	62	Polypropylene	Centrifugation (15mins) (3000rpm) Ultrafiltration (Amicon 30000 MWCO paper)
Yule 1994	Batch - Periodic shaking at specific intervals	1:50	68	Plastic	Centrifugation, Filtration (Amicon 30000MWCO paper)
Haigh & Hooker 1988	Batch - triplicate samples continuously rotated	1:10	7	Polypropylene	Centrifugation (15mins) (4100rpm)
Bell & Bates 1987	Batch - duplicate samples shaken in water bath	1:10	14	Polythene	Decantation & Centrifugation at 2500rpm (10mins)
McKinley and West 1981	Batch - Samples continuously agitated	DNA	DNA	Polypropylene	Centrifugation
Haigh et al 1989	Batch - duplicate samples shaken in water bath	1:10	14	Polyethylene	Centrifugation (4100rpm) (10mins)
Berry et al 1990	5 samples equilibrated in Gallenkamp shaker	1:10	70-112	Polypropylene	Centrifugation (15-20mins) (1500g)

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