LLW Repository Ltd

Low Level Waste Repository

LLWR Lifetime Plan

Inventory and near field

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10003 LLWR LTP Volume 3 Issue 01

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- the document demonstrates that the project is using the latest approved data; and
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HISTORY SHEET

10003 LLWR LTP Volume 3

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Inventory and Near Field

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Executive Summary

The Low Level Waste Repository (LLWR) is the UK's principal facility for the disposal of solid low-level waste (LLW). The site is owned by the Nuclear Decommissioning Authority (NDA) and operated on behalf of the NDA by a Site Licence Company (SLC).

The previous SLC, British Nuclear Fuels plc (BNFL), completed operational and post-closure safety cases for the LLWR in 2002. These were reviewed by the EA and, following a period of consultation, a new single authorisation was granted. The authorisation, with an effective date of 1 May 2006, is split into a number of schedules, of which Schedule 9 is a list of improvements and additional information that the operator must supply. Following this, the LLWR initiated the Lifetime Project, a programme of work aimed at addressing the requirements associated with Schedule 9 of the authorisation. The Lifetime Project encompasses a range of work areas, which will ultimately lead to the production of an updated Environmental Safety Case (ESC) by 2011 at the latest.

The present document is Volume 3 of a set of 5 volumes, with supporting references, that is designed to satisfy Requirement 2 of Schedule 9:

- Volume 1: Managing existing liabilities and future disposals at the LLWR;
- Volume 2: Assessment of Options for Reducing Future Impacts from the LLWR;
- Volume 3: Inventory and near field;
- Volume 4: Site understanding; and
- Volume 5: Performance update for the LLWR.

Volume 1 provides an overview of our submission and Volumes 2 to 5 provide more detailed information and analysis in support of Volume 1.

This document presents an overview of the work undertaken within the LLWR Lifetime Project focussed on the inventory and near field, and how our understanding of the near field system has advanced since the submission of the 2002 Operational and Post-Closure Safety Cases.

In the context of the LLWR, the near field incorporates the disposed inventory, the engineering features of the disposal system and that part of the natural environmental that is disturbed as a result of the construction, operation and closure of the facility. The disposed inventory itself is the sum of the activities disposed at the time of disposal.

The 2002 Operational and Post-Closure Safety Cases represented a significant body of work and the understanding gained of the evolution of the near field system, underpinned by an extensive programme of site monitoring, modelling and experimental studies, remains a source of information and conceptual models that are still valid within the current LLWR Lifetime Project. In terms of the inventory and near field, the LLWR Lifetime Project has focussed on improving this understanding, particularly in addressing issues highlighted within the EA's review and within our own assessment of the key uncertainties. In particular, the work has focussed on the development of a more robust disposal inventory, with an associated increase in understanding of waste heterogeneity, further understanding of the

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behaviour of key radionuclides within the system and the representation of interactions between waste components. Additionally, work has been undertaken to develop a greater understanding of the post-closure engineering system and its role in controlling the flow of water into and out of the near field.

Significant progress has been made in the derivation of an updated inventory for the LLWR site, particularly for historical disposals to the trenches. Much greater reliance has been placed on the use of real disposal records rather than the application of backfitting techniques to relate the historical disposals to current waste streams. This has resulted in a good knowledge of the inventories of key radionuclides and, importantly, a detailed understanding of the distribution of this inventory across the disposal system. For example, the representation of heterogeneity within the trenches is to the level of sub-bays. This information is important for subsequent modelling and the management of potential optioneering solutions. The inventory re-evaluation has also resulted in a reduction in the overall trench radionuclide inventory by around 20% from that calculated for the 2002 Safety Cases.

These improvements have been evident in the re-evaluation of the potential impacts of disposed uranium within the trenches, key radionuclides within the 2002 PCSC. The re-evaluation of the inventory of disposals at the LLWR, utilising historical disposal records, has resulted in a reduction in the trench uranium by a factor of two, with the vast majority (over 95%) of this uranium inventory associated with disposals from the Springfields site. The nature of this waste has been evaluated in detail and identified as process residues, with highly insoluble uranium bound within the residue matrix. A detailed geochemical model, supported by site observations, has been developed to represent the release of uranium from this waste form, which has shown that the uranium fluxes from the trenches calculated as part of the 2002 PCSC are conservative. As a result of this more realistic conceptual model of uranium waste behaviour and a representation of small scale heterogeneities, the rate of release of the majority of uranium from the trenches could be several orders of magnitude lower than previously modelled.

Geochemical modelling has also confirmed that the alkaline conditions associated with the vault waste form will persist for several thousand years.

A detailed evaluation of the performance of the near field engineering has also been undertaken. The assessment timeframes considered within the Lifetime Project are much shorter than those within the 2002 Post-Closure Safety Case, given the disruption of the wastes by coastal erosion around 5000 years After Present (AP). An assessment of the performance of the post closure engineering features, including cap, vertical drains and cut-off wall, has indicated that they will continue to function over the time period of interest and therefore play a significant role in limiting flows into and out of the facility.

As a result of this engineering performance assessment, a near field water balance model has been produced to represent the flow of groundwater through the near field environment, taking into account the site scale groundwater model and the behaviour and properties of the near field post closure engineering.

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This model indicates that, in general, vertical flows out of the near-field dominate, with the only significant horizontal flow being the flow from Vault 8 to the north-west. However, within the range of parameter uncertainties associated with modelled groundwater heads and trench base performance, there remains the possibility of additional horizontal flows from the trenches. The model also indicates that parts of the near-field environment, particularly the trenches, are likely to be at least partially unsaturated, although again this conclusion is very sensitive to the parameter uncertainties noted above.

This information has been combined with the key findings from the 2002 PCSC to produce an updated conceptual model of the near field. This conceptual model represents the main features, events and processes (FEPS) associated with the release of radionuclides from the near field environment. For the groundwater pathway, these have included inputs and outputs of water through the near field, controlled by the performance of the closure engineering, and the release of radionuclides, controlled by retardation processes and solubility limitations. A generally cautious approach has been adopted, particularly in relation to the leaching of waste into water. With the exception of uranium in the trenches, where this is good evidence for release being limited by the behaviour of the waste form, it is assumed that groundwater is in equilibrium with the radionuclide content of the waste, which is a cautious approach given parts of the facility are likely to be partially unsaturated.

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1. Introduction

The Low Level Waste Repository at Drigg (the LLWR) is the UK's principal facility for the disposal of solid low-level radioactive waste (LLW). The site is owned by the Nuclear Decommissioning Authority (NDA) and operated on behalf of the NDA by a Site Licence Company (SLC). United Kingdom Nuclear Waste Management (UKNWM) Ltd. holds a contract from the NDA for the management and operation of the LLWR and shares in the SLC were transferred to UKNWM Ltd on 1st April 2008.

To dispose of radioactive waste the SLC requires an authorisation under the Radioactive Substance Act of 1993 (RSA93), which under amendments of 1996 is determined by the Environment Agency (EA) for facilities in England and Wales. The LLWR has operated under the terms of authorisations under RSA93 (and previously under RSA60); the authorisations are periodically reviewed and renewed by the regulatory agencies.

Before granting or renewing an authorisation, the EA must satisfy itself that the proposed disposals are consistent with Government policy, for example that radioactive wastes should be managed and disposed in ways to protect the public, work force and the environment (HM Government, 1995), and are in accord with its own guidance on requirements for authorisation (Environment Agencies, 1997). This may include consultation with other agencies as needed.

It is the duty of the SLC to make submissions to the EA concerning the management and safety of the facility, and provide all necessary evidence on which the EA can make its determination. Provided it is satisfied, the EA may then grant an authorisation, which may be subject to conditions and requirements concerning both the operation of the LLWR and future actions and submissions by the SLC.

The previous SLC, British Nuclear Fuels plc (BNFL), completed operational and post-closure safety cases (2002 OESC and 2002 PCSC) for the LLWR in 2002 (BNFL, 2002a; 2002b). These were reviewed by the EA, and following a period of consultation, a new single authorisation was granted (Environment Agency, 2006a), encompassing all aspects regulated by the EA under RSA93. The authorisation, with an effective date of 1st May 2006, is split into a number of schedules, of which Schedule 9 (see Appendix A of Volume 1) is a list of improvements and additional information that the operator must supply.

We (the SLC) have initiated a programme of work – the LLWR Lifetime Project – to address the requirements of Schedule 9. The present document is Volume 5 of a set of 5 volumes, with supporting references, that is designed to satisfy Requirement 2 of Schedule 9:

Volume 1: Managing existing liabilities and future disposals at the LLWR;
Volume 2: Assessment of options for reducing future impacts from the LLWR;
Volume 3: Inventory and near field;
Volume 4: Site understanding;
Volume 5: Performance update for the LLWR.

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Volume 1 provides an overview of our submission and includes (Section 2, Volume 1) our approach to addressing Requirement 2 of Schedule 9; Volumes 2 to 5 provide more detailed information and analysis in support of Volume 1. The key deliverables provided in these reports comprise:

- information to show that our work is founded on national and international best practice (see Section 3 of Volume 1);
- assessment of options, to identify and assess the ways of managing the long-term impact of the Trenches (see Section 7 of Volume 1 and Volume 2);
- an analysis of the total 'radiological capacity' of the Vaults (see Section 6 of Volume 1);
- an updated view of site characteristics, the evolution of the engineered barriers and long-term performance, to support the previous analyses (Sections 4 and 5 of Volume 1 and Volumes 3, 4 and 5).

The submission focuses on the achievement and evaluation of post-closure safety consistent with satisfying Requirement 2 of Schedule 9; operational safety is not considered.

This volume – Volume 3: Inventory and near field –presents an overview of the work undertaken within the LLWR Lifetime Project focussed on the inventory and near field. The document presents key understanding and information that has been used to address Requirement 2, in particular to underpin the series of radiological calculations described in Volume 5. These calculations provide estimates of future impacts that are most pertinent to assessing the long term impacts of the options for future management of the LLWR. These have included calculations relevant to illustrating and quantifying the arguments within our submission concerning the radiological capacity and future management strategy for the LLWR.

A particular focus of this volume is demonstration of how our understanding of the near field system has advanced since the submission of the 2002 Operational and Post-Closure Safety Cases. The 2002 Operational and Post-Closure Safety Cases represented a significant body of work and the understanding gained of the evolution of the near field system, underpinned by an extensive programme of site monitoring, modelling and experimental studies, remains a source of information and conceptual models that are still valid within the current LLWR Lifetime Project. In terms of the inventory and near field, the LLWR Lifetime Project has focussed on improving this understanding, particularly in addressing issues highlighted within the EA's review and within our own assessment of the key uncertainties.

The document is structured as follows:

Section 2 presents an overview of the near field environment at the LLWR, including a description of the site, nature of the waste and the engineered features.

Section 3 provides a summary of the output from the 2002 PCSC, focussing on the evolution of the biogeochemical conditions within the near field environment.

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Section 4 provides an overview of how the near field inventory has been updated since 2002, with a particular focus on historical disposal records and the heterogeneity of the system

Section 5 is concerned with the biogeochemical modelling that has been undertaken within the current LLWR Lifetime Project. Modelling of the behaviour of uranium provides the focus of this work.

Section 6 summarises the engineering performance assessment, including a near field water balance model.

Section 7 presents the overall conceptual model of the near field, based on the sections above and the key lessons from the 2002 PCSC.

1.1 The role of the near field in the 2002 Post-Closure Safety Case

The 2002 Post-Closure Safety Case (PCSC) was developed through a systematic treatment of all features, events and processes (FEPs) relevant to the LLWR disposal system, both now and in the future, aimed at identifying the potential importance of uncertainties associated with this understanding (BNFL, 2002b). This required consideration of the conceptual and physical basis of models used to represent the system and the algorithms employed in the mathematical codes.

Within the 2002 PCSC, a wide range of assessment calculations were undertaken, encompassing the groundwater pathway, disruption of the site through coastal erosion processes and future human actions. For all of these, the near field provided the source term for subsequent assessment calculations.

The near field environment also presents a number of barriers to release of radionuclides from the disposed inventory to the accessible environment. The first of these is physical containment, represented by the design of the trenches and vaults and by the closure engineering, such as the proposed cap and cut off wall. These act to control the flow of water into and out of the near field environment and so restrict the flux of radionuclides into the geosphere. Additionally, these engineering features can also restrict the flow of radionuclides in gaseous form or limit the extent of human intrusion. The near field environment also presents a chemical barrier to the release of radionuclides. The mobility of many radionuclides is dependent on the ambient geochemical conditions encountered within the near field environment. The establishment of beneficial geochemical conditions, for example high pH or an anaerobic environment, can limit the release of radionuclides from the near field.

The basis of the near field work within the 2002 PCSC was therefore the development of a sound knowledge of these potential barriers and to gain a good understanding of the behaviour and evolution of the disposal environment, to provide, for example, radionuclide fluxes into the geosphere or as the basis for the assessment of future human actions. The key components of this understanding were:

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- An understanding of the disposed inventory, covering disposals from the beginning of operations through to final closure;
- Engineering features and groundwater flow within the near field environment;
- The evolution of the biogeochemical conditions within the disposal system; and
- The release and transport of radionuclides.

Ultimately, the near field was described through the application of the biogeochemical model, DRINK (BNFL, 2002c), which has the capability to represent the key mechanisms controlling the behaviour of radionuclides. Within the 2002 PCSC, these processes included waste degradation, microbial reactions, geochemical buffering reactions, groundwater flow, radionuclide solubility and sorption, corrosion and radioactive decay.

A key part of the 2002 PCSC was the demonstration of good science and engineering, as required by the EA's guidance on requirements for authorisation (Environment Agency *et al*, 1997). Consequently, the biogeochemical near field model was supported by a wide range of experimental and underpinning modelling studies, aimed at gaining a more robust understanding of the key processes operating within the near field environment.

1.2 Environment Agency review of near field aspects of the 2002 Safety Cases

Following submission of the 2002 Post-Closure and Operational ('Environmental') Safety Cases for the LLWR, the EA undertook a thorough and detailed review which led to the publication of its assessment of the Safety Cases (Environment Agency, 2005b). This process provided key inputs into the EA's subsequent review of the Authorisation for the facility. The Authorisation review culminated in proposed updates to the site Authorisation as outlined in the Explanatory Document (Environment Agency, 2005a), and following public consultation the final Authorisation as described in the Decision Document (Environment Agency, 2005b) and the Compilation of Environment Agency Requirements (Environment Agency, 2006).

The assessment was underpinned by a very detailed study of the Safety Cases, undertaken by a range of expert Review Groups each charged with assessing a particular technical area of the submissions. These were coordinated by a Core Review Group which produced the overall, high-level findings of the assessment. All review issues and the assessment thereof were documented in a thorough and detailed manner, resulting in the production of a large number of Issue Assessment Forms (IAFs), provided to the LLWR SLC in the form of the EA's Issues Database. There are 132 IAFs in total, each of which considers a particular aspect of the Safety Cases, summarises the EA's views and typically contains several recommendations.
These IAFs have been actively managed during the current LLWR Lifetime Project. The scope of the Lifetime Project has been established based on the key issues that influence the environmental safety of the site (Randall *et al.*, 2006; Lean and Fowler, 2007), including addressing the IAFs. In particular, six monthly reviews of the IAFs are undertaken against progress within the LLWR project (e.g. Lean, 2007).

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The EA's review considered that the near field modelling was a reasonable illustration of the evolution of the waste and the release of radionuclides (Environment Agency, 2005b). Indeed, the EA's reviews of the Nirex disposal concept (Environment Agency, 2007) noted how the approaches adopted by the LLWR to examine spatial and temporal heterogeneity in near-field biogeochemistry could be applied to geological repositories in future.

Nevertheless, a number of IAFs were produced by the EA that were relevant to near field aspects of the 2002 PCSC. Together, with the reviews and scopes of work recorded in Randall *et al*. (2006) and Lean and Fowler (2007), highlighted a number of key issues to be focussed upon, namely:

- The development of a more robust inventory through greater use of historical data rather than a reliance on the backfitting of current disposal waste streams;
- A greater understanding of waste heterogeneity;
- Addressing the key uncertainties within the near field representation, such as interactions between waste components and the role of organics and colloids;
- A further understanding of the behaviour of uranium, the key radionuclide within the 2002 PCSC;
- Issues associated with representing heterogeneities within the system; and
- Greater understanding of the engineering system and the effect on near field flows.

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2. Design of the LLWR

2.1 Description of the near field

In terms of an analysis of the impacts resulting from the release of radionuclides from the LLWR facility, the near field of the LLWR site represents the source component of the source – pathway – receptor system. The near field incorporates the disposed waste, the engineered features of the disposal system and that part of the natural environment that is disturbed as a result of the construction, operation and closure of the facility. The spatial extent of the near field extends to the limits of the final cap. In practical terms, the top of the near-field is defined by the upper boundary of the intrusion barrier within the final cap, since the soil zone overlying the cap is within the biosphere. The lower boundary of the near-field corresponds to the base of the disposal facilities and the proposed cut-off walls and vertical drain.

The LLWR has been the principal facility in the UK for the disposal of low-level waste (LLW) since 1959. The LLWR receives waste from a range of consignors, including nuclear power stations, fuel cycle facilities, defence establishments, general industry, isotope manufacture sites, hospitals, universities and the from the clean up of historically contaminated sites.

Two distinct disposal systems are present within the LLWR site. From 1959 until 1995, waste was tumble tipped into pre-excavated trenches (see Figure 2.1), in the north-east corner of the site. The base of each trench was generally excavated into a low permeability clay-rich layer of the natural geology at a depth of 5 – 8m below ground level. No compaction of the waste was undertaken other than that which occurred through the weight of the overlying waste and cover layer. A total volume of approximately 800,000 m^3 of waste has been deposited in the trenches.

Since 1988, waste has been disposed into an engineered, concrete disposal vault, Vault 8 (Figure 2.1). This upgrade to the disposal operations was aimed at making more efficient use of space and to improve the visual impact of disposal operations. At the same time, remedial work was commenced on the trenches, including the installation of a low-permeability cut-off wall (to limit lateral movement of groundwater and radionuclides), interim capping of the filled trenches and refurbishment of the leachate drainage system.

The vault is designed to take grouted ISO containers and also items of waste that are too large for a container. With a total capacity of about 200,000 m^3, Vault 8 is now approaching capacity and further vaults are required in order to meet future requirements. The current plan is for the construction of seven additional vaults (Vaults 9 to 15: referred to as “future vaults”) which will allow storage and disposal operations to continue to at least 2050.

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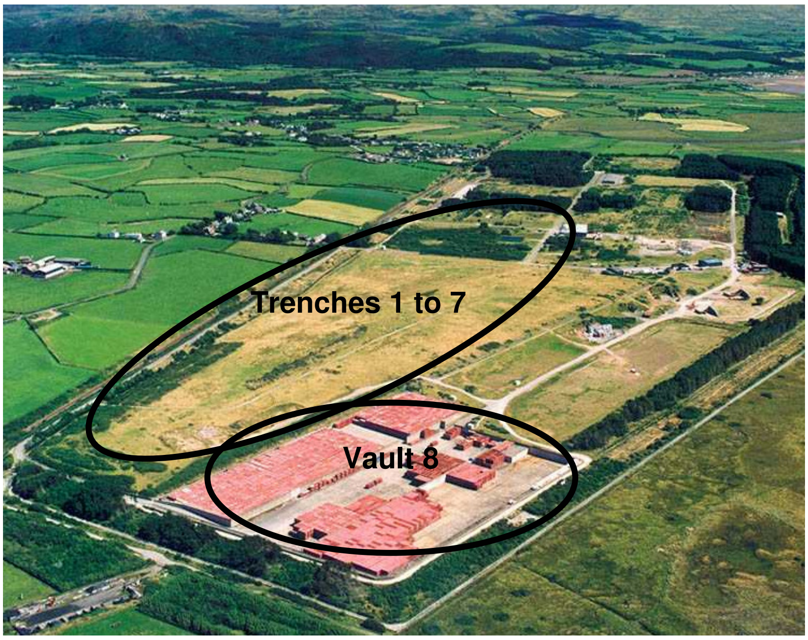

Figure 2.1 Aerial photograph of the LLWR with Trenches 1 – 7 and Vault 8 highlighted (view from north-west to south-east)

2.2 Wastes and wasteforms

The wastes disposed to both the vaults and the trenches have broadly similar distributions of waste materials being comprised principally of organics, including cellulose and plastic, metals, contaminated soils and inorganics, including silicates, fluoride residues and thorium ore minerals. The characteristics of the waste materials are discussed further in Section 3.

The major difference between the trenches and the vaults is associated with the waste form. The disposal technique for the trenches involved the loose tipping of generally unconditioned waste (typically in plastic bags and mild steel drums) into unlined trenches. Other waste items were disposed of directly to the trenches; these included scaffolding planks, pipes, packaging materials, pallets and redundant laboratory and office equipment. No mechanical compaction was undertaken, except that which occurred naturally as a result of the placement of wastes; some self-compaction and settlement has taken place since disposal. The trenches were filled with waste to within about a metre of the ground surface and were topped off with about 1m of backfill (1.5m in Trench 7 from 1988 onwards). The backfill was principally derived from material obtained as a result of the excavation of the trenches.

Within the vaults, wastes are containerised in steel overpack International Standards Organisation (ISO) containers, with the voidage in the ISO containers filled with a low viscosity cementitious grout prior to disposal. The grouted ISO containers thus provide an essentially monolithic waste form, which is then emplaced within the vaults using conventional large capacity fork-lift trucks.

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The disposal of some large items of waste in the vaults is impractical using the preferred steel containers. Allowance has therefore been made in the designs for the in situ grouting of such items into prepared areas of the vault using mobile grouting facilities.

Once a vault has been completed any significant residual void space around the containers will be in-filled with coarse granular inert material, such as a crushed rock aggregate. Compared with the trenches, which are essentially voids filled with unconditioned wastes, the vaults contain a substantial volume of additional material (steel and cementitious materials) that has been added to the waste as part of the containerisation and grouting process.

2.3 Near field engineering

At present, the site is in an "operational phase", whereby disposal activities are on-going. It is predicted that this phase will continue until 2050. After this date, the site will undergo a "management phase" where emplacement of engineering structures associated with site closure will be installed. Following this, the LLWR will enter a "post-closure" phase, following withdrawal of control over the facility by the operator.

These proposed site development operations will alter the existing engineering structures at the site and add new features.

The main engineering features currently present at the LLWR site include:

- The trenches;
- The interim cap;
- The cement bentonite cut-off wall (around the north-east and north-west boundaries of the trenches);
- The leachate management system; and
- Vault 8.

In terms of the future engineering features of the site, to be present at the LLWR following the withdrawal of control of the facility, a design option termed the Single Option has been developed (with full details to be found in Carpenter and Proctor, 2007; Belton, 2007). The Single Option is a holistic option that describes current and future site management (including both the vaults and the trenches) and closure features (e.g. cap and cut-off wall) that are consistent with the proposed design concept. The main features of the Single Option design include:

- The trenches;
- Vault 8;

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- Vault 9 and the proposed future vaults;
- The proposed final cap;
- The proposed cut-off wall; and
- The proposed vertical drain.

Schematics of the near-field engineering, identifying key features and their spatial relationship, are provided in Figures 2.2 and 2.3. The Single Option design is intended to be a multi-barrier design concept and aims to minimise the amount of leachate, as far as practicable, by reducing infiltration and contain any leachate generated for as long as reasonably achievable.

The option includes an impermeable cap, which is characterised by two domes over the trenches and vaults with a central valley between these domes, and the incorporation of bio-intrusion, vegetative, barrier, drainage and gas control elements. Beneath the vaults is a double liner system to provide leachate retention for as long as possible.

A vertical drain, which can be installed at any time up to the end of the management phase, is designed to protect the Upper groundwater system (Volume 4) from discharge of leachate. Leachate could eventually overtop the north-east walls of the vaults (which are lower than the other walls) and flow into the vertical drain, which will extend downwards into the Regional groundwater (Volume 4).

A cut-off wall is provided to minimise the movement of groundwater between the disposal area and the Upper groundwater system. As such, the cut-off wall may be taken to provide a second line of defence should the vertical drain and/or the vault liner system cease to function adequately (the trenches do not have a liner system as such as the base of the trenches is a natural, clay-rich glacial sediment augmented with bentonite).

The overall concept and the materials used for the main engineering components of the Single Option are similar to those of the 2002 PCSC, with the following key differences:

- Cap profile: the design of the Single Option cap, comprising two domes, is significantly different from the 2002 PCSC design, which assumed an asymmetrical dome with falls to the perimeter.
- Vault base liner: the Single Option has a higher specification liner system.
- Vault walls: the Single Option has a higher design specification for vault walls incorporating a bentonite enhanced soil (BES) layer for increased hydraulic performance.
- Vertical drains: these are optional and of different design (location, type and plan area) in the Single Option.
- Control of leachate discharges post-closure: The Single Option allows filling up of vaults and controlled overtopping of the wall between the vaults and trenches.
- Different drainage arrangements for Vault 8 and future vaults during the operational phase.

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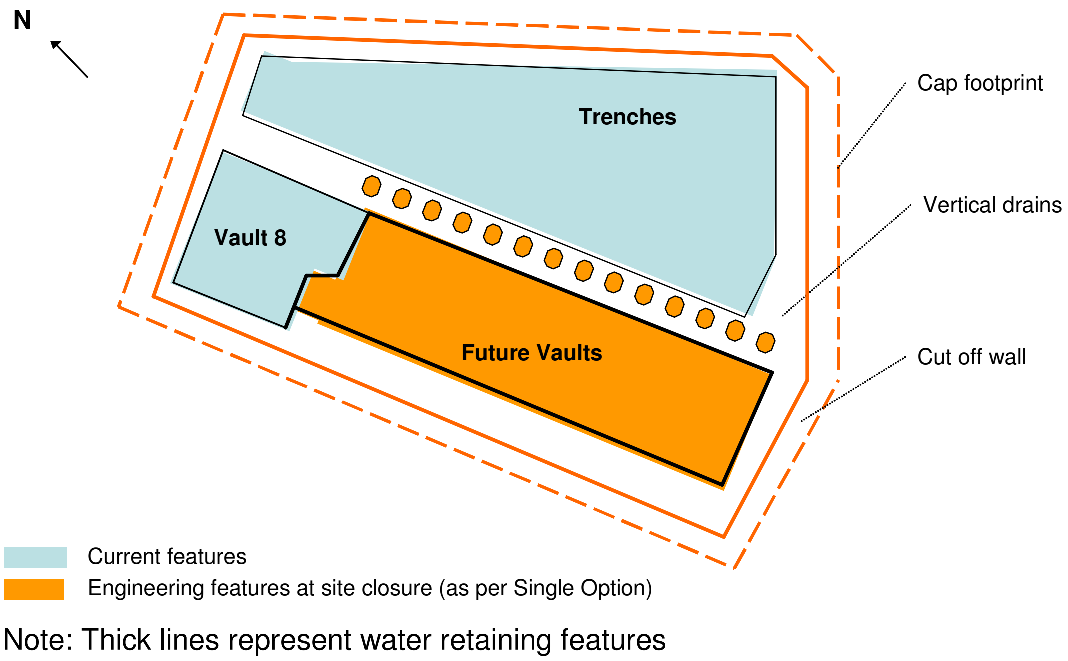

Figure 2.2 Plan view of proposed main engineering features within the LLWR disposal area following withdrawal of control (not to scale).

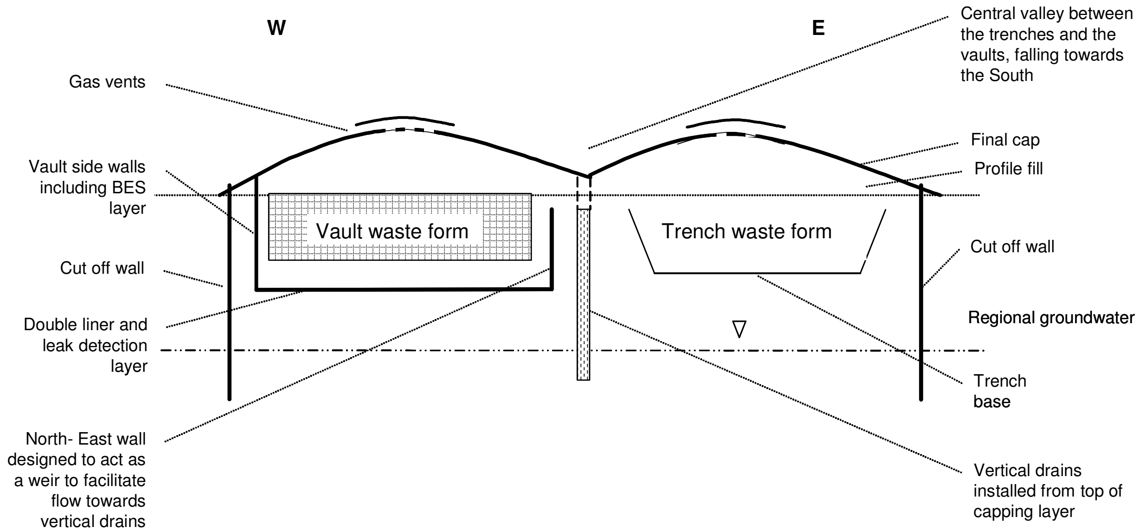

Figure 2.3 Cross-section through the proposed main engineering features within the LLWR disposal area following withdrawal of control (E-W cross section through Future Vaults) (not to scale).

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3 2002 Post-Closure Safety Case Near Field and Inventory

3.1 Introduction

In the 2002 PCSC, a significant integrated programme to develop a robust and scientifically sound representation of the near field environment of the LLWR site was undertaken. Much of this work has also been published in the open, peer reviewed literature (e.g. Small *et al.,* 2000, 2004; Beadle *et al*., 2001, 2003), as well as being reported in the 2002 PCSC (e.g. BNFL, 2002c,d,e).

This section provides a summary of the key aspects of the near field within the 2002 PCSC. Many of these aspects, particularly those related to the biogeochemical evolution of the site, remain appropriate for the current radiological safety assessment. Subsequent sections of this report describe how this knowledge has been updated and enhanced since the 2002 PCSC.

3.2 The inventory

The radionuclide and waste materials inventory information used in the 2002 PCSC was based on disposal records and the most recent available UK National Inventory for 1998, which characterised the various LLW streams and the stocks and arisings of raw wastes, both from existing facilities from prospective facilities. The inventory is based upon assumptions about the relationship between volumes and types of raw wastes and the disposal of the conditioned wastes derived from them.

Drigg inventory information was derived for three different time periods, pre 1993, 1993-98 and post 1998 depending on the information sources available (BNFL, 2002e). For the period up to the end of June 1993, the past disposal records were used as the source of data on waste volumes and the 1998 UK National Inventory was used as the source of detailed radionuclide and material contents of the waste. This involved assigning an association between the appropriate waste streams in the UK National Inventory and the real waste disposed in the past – a “backfitting” process. For the period from the start of July 1993 to the end of March 1998, a LLWR Database was used as the source of weight and radionuclide information, with the 1998 UK National Inventory used as the source of material contents. By this time, the LLWR Database was recording the waste streams from which each disposal consignment arose. For the period from the start of April 1998 to the projected end of disposals, the 1998 UK National Inventory was used as the source of all waste information.

The inventory derivation, and the assumptions that underpin it, is explained in great detail in the inventory of disposals report (BNFL, 2002e). The inventory of disposals report also covered the issues of uncertainty and spatial heterogeneity. It was assumed that the trench and vault radionuclide contents as provided in the inventory were all present at the end of operations, with no allowance for decay, in-growth and possible leaching in the period between disposal and site closure. This was justified on the basis that the highest risks were expected to occur with the longer-lived radionuclide species given the extended timescales that were considered during the 2002 PCSC and so any decay between the time of disposal and the end of operations in 2050 was assumed to have a negligible effect on the PCSC results.

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3.3 Near field biogeochemical evolution

Biogeochemical processes operating in the near field control the evolution of the local environment of disposed radionuclides in LLW. In turn, the chemical environment influences the mobility of many radionuclides in groundwater and also is important to processes that control the release of radionuclides from solid waste to the aqueous and gaseous phases.

Consequently, an understanding of these processes was a key driver for the near field work performed within the 2002 PCSC. The work was undertaken using the biogeochemical code DRINK and reported in BNFL (2002c), which also contains a review of the key biogeochemical processes which may influence radionuclide release and mobility. These processes are summarised below.

As a consequence of the nature and chemical composition of the organic (cellulosic) and metallic (iron) containing LLW, the principal degradation process will involve oxidation. It is well established that microbial processes mediate and catalyse these slow waste degradation processes, particularly of organic materials. Iron corrosion processes are recognised to occur spontaneously, however products of corrosion such as hydrogen may be readily utilised by microbes and microbial action can lead to enhanced rates of corrosion. Cementitious materials comprising the Drigg vault waste form may result in the establishment of alkaline chemical conditions, which may affect the degradation of organic matter and corrosion in the Drigg vault environments. Understanding the processes of waste degradation is important to characterising the chemical environment of radionuclides in LLW over various timescales. Site monitoring studies and waste simulation studies have been undertaken to support the development of this understanding. These are described in full in BNFL (2002c).

The representation of these processes was undertaken through the DRINK biogeochemical code, the functionality of which includes groundwater flow, radioactive decay, waste degradation, microbial reactions, sorption, gas generation and geochemistry. Incorporation of all these processes within one model enabled the evolution of the near field to be simulated over the assessment time period and the release of radionuclides from the near field to be calculated.

A key output of the model was the simulation of the evolution of the ambient geochemical conditions within the near field environment. For example, Figures 3.1 and Figure 3.2 show summary results of the near field model, in terms of pH and redox (pe) development. The principal effects of the disposal of waste and waste form materials in the trench and vault environments were to produce reducing conditions by processes of organic waste degradation and corrosion. The model predicted the establishment of chemical conditions in the trenches that were significantly more reduced than in the Drigg geosphere. Site monitoring confirms that reducing conditions are already being established in the trenches, characterised by anaerobic conditions and the production of reduced chemical species such as methane, acetate and sulphide (BNFL, 2002c).

Furthermore, reducing conditions were simulated to occur in the trenches for a period up to around 2,500 to 4,000 years. The duration of this period of reducing conditions was predicted to vary across the trench system, principally due to the heterogeneity in the inventory of LLW

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materials. Model sensitivity studies also indicated that the duration of this period of reducing conditions was dependant on the flow of groundwater through the near-field and by the concentration of oxidant species (nitrate, dissolved oxygen) in groundwater.

Acidic conditions were predicted to develop in the trenches as a result of the production of acetate and carbon dioxide resulting from the degradation of cellulose by the microbial processes included in DRINK. After around 2,000 years, when cellulose was predicted to be fully degraded, pH increased above that of the groundwater; this is a result of iron corrosion processes which yield hydrogen and generates alkalinity. Thus, pH varies in the model as the relative rates of cellulose degradation and corrosion change with time.

In the vaults, alkaline conditions were predicted to develop as a result of the presence of a large proportion of cementitious material releasing alkalis to groundwater. Evidence for the establishment of reducing and alkaline conditions in the vault environment comes from experimental simulant studies (BNFL, 2002c). As shown in Figure 3.2, the DRINK model predicts that the dissolution of this cementitious material would result in these alkaline conditions being maintained for many thousands of years

The behaviour of disposed radionuclides is frequently dependent on the prevailing chemical conditions, and the DRINK model reproduced these features. In particular, the aqueous concentration of uranium was shown to be strongly influenced by the varying chemistry through its effect on uranium solubility. During the initial period of reducing conditions uranium was solubility limited by the uranium (IV) amorphous oxide UO_2. The solubility of this phase varied in the range 1E-4 and 1E-8 mol l^{-1} as pe and pH vary. After 4,000 years, once more oxidising groundwater conditions were re-established, uranium (VI) was stabilised, resulting in much increased solubility and uranium concentrations above 1E-4 mol l^{-1}. At this high concentration uranium was washed out of the near-field limited by sorption processes. The release of uranium, specifically uranium-234, from the trenches upon reoxidation was a key process determining the peak radiological dose for the groundwater pathway in the 2002 PCSC (BNFL, 2002b), with the dose principally due to the decay products of uranium, particularly Pb-210.

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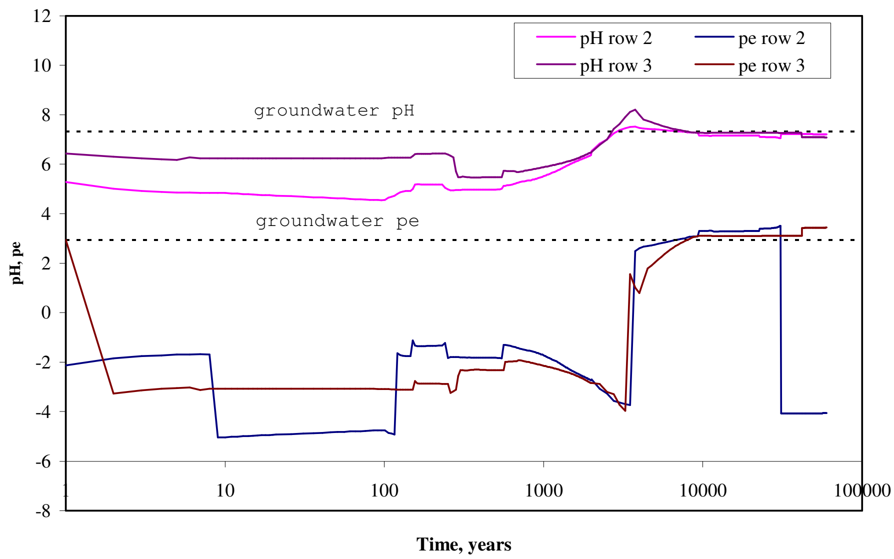

Figure 3.1 Modelled pH and pe in representative trench cells within the DRINK model

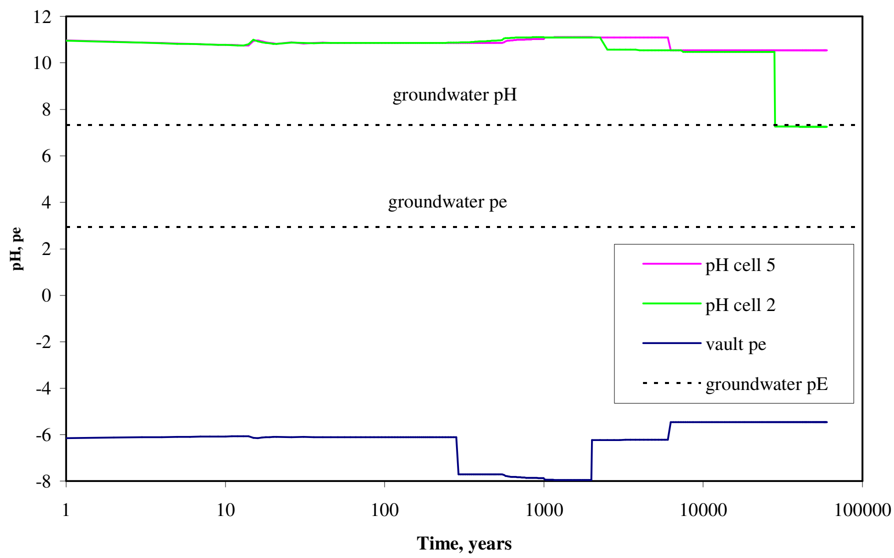

Figure 3.2 Modelled pH and pe in representative vault cells within the DRINK model

The understanding gained during the 2002 PCSC of the biogeochemical evolution of the near field system represents a source of information and conceptual models that are still valid within the current LLWR Lifetime project. In particular, the underpinning information used to develop and parameterise the DRINK near field model provides a framework from which the current radiological assessment can be built. It is recognised that the understanding of the site has evolved since 2002, particularly in relation to groundwater flow (see Volume 4), inventory (Chapter 4 of this volume), uranium waste form (Chapter 5 of this volume) and engineering performance (Chapter 6 of this volume). All of these will influence the evolution of the near

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field system to some extent. For example, the timing of any re-oxidation of the trenches will be dependent on the flow of groundwater through the near field, which in turn is controlled by the performance of the engineering system.

Additionally, it is now expected that coastal erosion processes will be more significant than assumed in 2002, with coastal erosion is expected to result in the destruction of the site between 750 and 2500 years AP, depending on the rate of sea-level rise. (see Volume 4). There is potential that this could impact on the evolution of the near field environment through changes to the hydrogeological regime resulting from these coastal erosion processes and landscape change. Also, as it is anticipated that the site will be destroyed by coastal processes within some thousands of years, there may not be time for the re-oxidation of the trenches to occur.

However, it is expected that the main features of the near field evolution, namely the establishment of reducing conditions through the degradation of organic waste and corrosion of metal and the development of high pH conditions within the vaults, remain valid for the current radiological safety assessment.

The subsequent sections describe how this information has been developed within the LLWR Lifetime Project, culminating in a refinement of the conceptual model for the near field described in Section 7.

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4 Updates to the Inventory

4.1 Introduction

Following the production of the 2002 Safety Cases, a number of areas of potential improvements to the derivation and assessment of the disposal inventory have been identified. These improvements were identified both from our own assessment of the 2002 Inventory (Wareing, 2004) and the issues and recommendations identified by the EA (Environment Agency, 2005). These latter are described in Section 2.

The key areas for improvement identified in these two separate reviews can be summarised as follows:

- Non-radiological aspects, in particular the characterisation of the physical form of specific disposals where radionuclide release models are dependant on waste form;
- Reduction in the uncertainty in the activities of key radionuclides identified by the Post-Closure Radiological Safety Assessment (PCRSA);
- Understanding of waste heterogeneity;
- The methodology for backfitting historical trench routine disposals from the1998 UK National Inventory data:
- The model used in the calculation of packaged waste volumes; and
- Greater use of historical documentation from plants consigning waste.

Specifically, the EA review concluded that the inventory could be made more robust through greater use of historical data rather than a reliance on the backfitting methodology, with a focus on the activity levels and heterogeneity of radionuclides with the greatest impact. Additionally, the work to derive the inventory could be more appropriately underpinned by a demonstration that all disposal records of significance have been assessed, coupled with a clear description of the methodology.

4.2 Methodology

The inventory calculated for the 2002 Safety Cases was derived from a combination of data extracted from disposal records and the 1998 National Inventory, the most up to date published source available at the time. The National Inventory was used to both predict future disposals to the Vaults and for backfitting inventory data from the historical disposals, particularly to the trenches. This backfitting exercise assumed that present day waste streams are similar to those disposed in the past and thus the detailed radionuclide fingerprints and material contents in the National Inventory can be applied to the historical waste volumes.

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For the production of the present disposal inventory, however, a much greater reliance has been placed on historical disposal records, supported by a range of other data sources. The sources used in deriving the information are:

- Operational Data: information on current and historical disposal practices, trench and vault dimensions and disposal container properties.

- Disposal Records: primary source for historical disposals. However, variability in reporting requirements over the years means that these need supplementing with other data sources.

- Waste Tracking System database: an electronic database capturing disposal records since the beginning of Vault 8 operations in 1998.

- 2004 UK National Inventory: inventory database (Electrowatt – Ekono, 2005) that lists radioactive waste by waste streams, each containing information on radiological composition and physical and chemical properties.

- D4 Index table database: an electronic database cataloguing the transfer of paper D4 disposal records to microfilm. The database lists information on consignor or building (for Sellafield wastes) of origin, location of paper-based records and date of disposal for each consignment.

- Summarised volumes: month end and period end reports recording volumes of waste for each consignor, and BNFL Annual Environmental Reports.

- Public Record Office documents: source of a number of reports containing supplementary information to that available in the National Inventory and disposal records, which have been used to provide waste activity and physical/chemical data for a number of key trench disposals.

- Lifetime Plan and waste accountancy data: over-arching documents, published annually, describing the activities required to take each NDA site to its agreed end state. These have been used principally in sensitivity analyses through providing scenarios for the selection of waste streams for the future vaults inventory calculations.

- Historical UK Radioactive Waste Inventories: although the current National Inventory contains more detailed information, the use of previous National Inventories is useful when backfitting historical disposals.

- Waste Inventory Disposition Route Assessment Model (WIDRAM): WIDRAM has been developed by Nexia Solutions on behalf of the NDA to hold comprehensive LLW waste inventory data. The data held within WIDRAM have been used to supplement the National Inventory information, particularly in the derivation of the future vaults inventory (Wareing, 2007).

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A full description of how these data sources are used to derive the disposal inventory of the LLWR site is given in Wareing *et al.* (2008, Sections 2 – 5). In general terms, the trenches inventory has been largely derived from the extensive use of actual disposal data, with the need for backfitting of contemporary waste stream data to historical disposals kept to a minimum. The inventory for Vault 8 has been calculated almost entirely from actual disposal data, with minimal support provided by the detailed radionuclide and material fingerprints from the 2004 UK National Inventory where necessary. The Vault 8 inventory is available by individual consignment, allowing a detailed investigation of waste heterogeneity. The methodology for the calculation of the future vaults inventory has also been improved from the 2002 Safety Cases. Although primarily informed by the 2004 National Inventory, the use of operational data available in the Waste Tracking System database means that the modelling of packaging factors in the future vaults is representative of current operational experience.

Table 4.1 summarises each data source used in deriving the inventory of disposals and shows for the trenches, Vault 8 and the future vaults whether each source has been used for direct derivation, backfitting, prediction or sensitivity analysis. The approximate order of priority for the use of the data sources for each of the trenches, Vault 8 and future vaults inventory calculations is given numerically in the coloured squares.

Table 4.1 Use of data sources in deriving the LLWR inventory of disposals.

	Trenches 1 to 7	Vault 8	Future Vaults
Operational Data	1	2	1
Historical Disposal Records	2		
LLWR Waste Tracking System database	7	1	4
D4 Index table database	3		
Public Records Office Reports	4		
Month-end reports and period-end reports	5		
BNFL Annual Environmental Reports	6		
2004 UK National Inventory	8	3	2
Historical National Inventories	9	4	
WIDRAM	10	5	3
NDA Lifetime Plan and waste accountancy			5

Key: Data used directly (green); Data used to 'backfit' (yellow); Data used to predict (cyan); Data used in sensitivity analysis (orange); Data not used (white)

4.3 Overview of the derived inventory

The disposals inventory has been calculated separately for each of three distinct disposal areas at the LLWR; the trenches, Vault 8 and Future Vaults, and has been reported in full in Wareing *et al.* (2008, Section 6) which includes a detailed analysis and description of individual radionuclides, non-radiological disposals and the impact of alternative scenarios for the management of UK LLW. These inventory databases have been combined into a single Microsoft Excel spreadsheet, which has provided a key component for other tasks within the Lifetime Project. These tasks have included the near field modelling reported in this Volume, the radiological risk assessment calculations presented in Volume 5 and the development of

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appropriate management strategies and an analysis of the total radiological capacity of the site (see Volume 2).

The derived inventory presented in Wareing *et al.* (2008) represents the radiological and non-radiological components of the waste and any associated materials such as waste containers and waste immobilisation matrix material. It does not include information on the structures of the vaults or trenches nor any material used to cover the waste after disposal. The radiological component of the inventory represents the sum total of the activities disposed to the site at the time of disposal. Changes to the inventory due to radioactive decay and in-growth and the leaching of radionuclides into the wider environmental over the previous fifty years of disposals at the site have not been represented in the current inventory.
.
A key feature of the inventory presented here is the much higher level of detail than was possible for the 2002 Safety Cases. In 2002, the trench inventory, for example, was derived per “mesh cell”, each of which represented approximately one seventh of a trench. In contrast, the improved methodology for the calculation of the historical inventory has made it possible to provide a trench disposal inventory at a much higher level of discretisation. Within the trenches, disposals were tumble-tipped into designated disposal bays, each of which was divided into four sub-bays (shown in Figure 4.1). The improved methodology for the calculation of the historical inventory, which makes use of the disposal dates available in the D4 index table database, has made it possible to provide a trench disposal inventory by trench sub-bay.

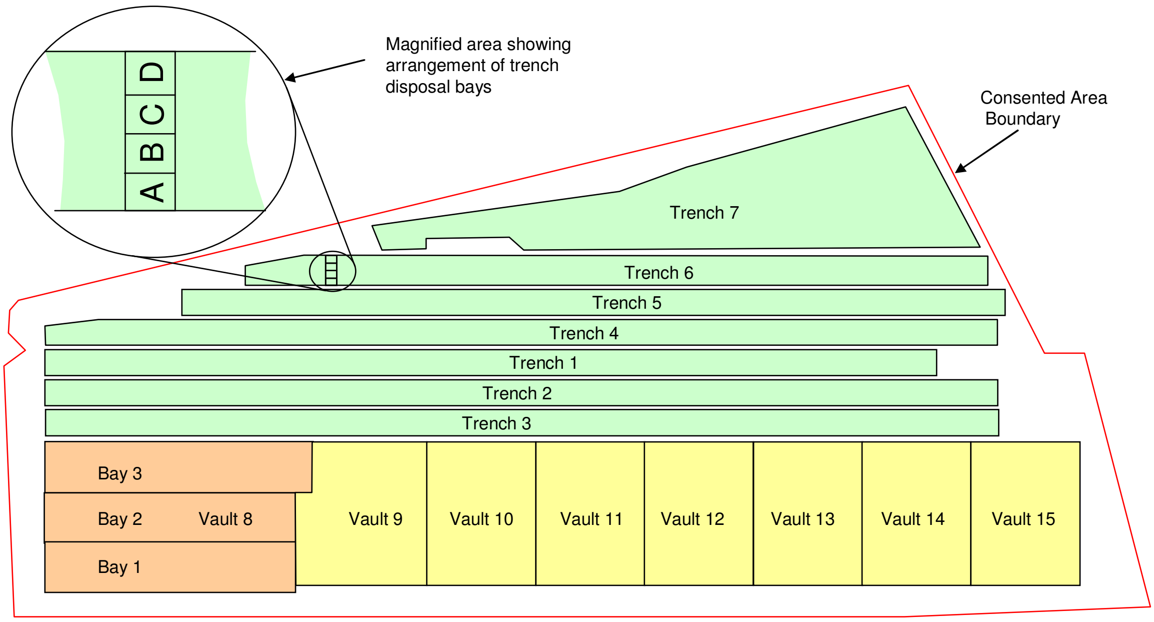

Figure 4.1 Schematic layout of the LLWR disposal areas

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Similarly, the use of the Waste Tracking System for the development of the Vault 8 inventory has enabled the inventory of each individual container to be determined. For the future disposal vaults, the use of the annual predictions of LLW arisings within the 2004 National Inventory has enabled the future arising inventory to be determined for each year up to the end of site operations.

The format of the inventory model therefore allows a level of detail significantly higher than that available previously. In particular, it is now possible to represent both the radiological content and materials properties of the waste at the level of individual sub-bays within the trenches. This is clearly important for subsequent modelling activities but also in relating decisions to risk management and identifying any potential targeted interventions as part of the site's management strategy (see Volume 2).

Section 6 of Wareing *et al.* (2008) presents the radionuclide and material content of the waste in terms of activity and volumes for each disposal trench, the existing Vault 8 and each of the future vaults 9 to 15. This indicates that the site is predicted to be volumetrically full by around 2055, with the volumes of waste disposed to the trenches approximately equal to that disposed to the vaults.

Figure 4.2 shows the distribution of radionuclides across the trenches, Vault 8 and Future Vaults, whilst Table 4.1 shows how the inventory derived as part of the Lifetime Project compares with that associated with the 2002 Safety Cases. Wareing *et al* (2008) present how the change factor was derived from inventories of each of the key nuclides. Overall, the total inventory has changed little from 2002 – for the key radionuclides presented in Table 4.2, the total inventory has dropped very slightly from 1,200 TBq in the 2002 Safety Cases inventory to 1,180 TBq in the 2008 Lifetime Project inventory.

There has, however, been a change in the relative proportion of activity between the trenches and the vaults. In 2002, the trench disposals accounted for around 75% of the total inventory, whereas within the Lifetime Project inventory, the trench disposals account for around 50% of the total. The trench inventory has reduced by around 20% from that calculated for the 2002 Safety Cases. Another notable change is in the Vault 8 inventory, which has reduced by a factor of two. These changes are primarily due to the greater use of actual data in calculating the trench and Vault 8 inventories. In contrast, the future vaults inventory has increased by a factor of 1.5, mainly due to the generally higher activities in the 2004 National Inventory when compared to the 1998 National Inventory, which was used in the calculation of the 2002 Safety Cases inventory.

Additionally, a number of specific radionuclides show significant differences between the current inventory and that derived in 2002, or are dominated by specific disposals. These are discussed in the following sections.

In terms of the non-radiological aspects of the inventory, Figure 4.3 shows the distribution of these materials within the trenches, Vault 8 and future vaults. The most prevalent waste materials predicted to arise in the combined inventory are ferrous metal, which is present at approximately the same proportions within the trenches, Vault 8 and future vaults. There is

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also a considerable content of a variety of plastics and cellulosic materials in both trench and vault wastes: these are common constituents of operational wastes. Additionally, significant quantities of concrete, rubble and soil are present arising from decommissioning wastes. This is most noticeable in the future vaults, where a large volume of soil is predicted to be disposed. This largely represents waste from contaminated land associated with site clean-up on NDA sites, where the disposal route is declared as the LLWR.

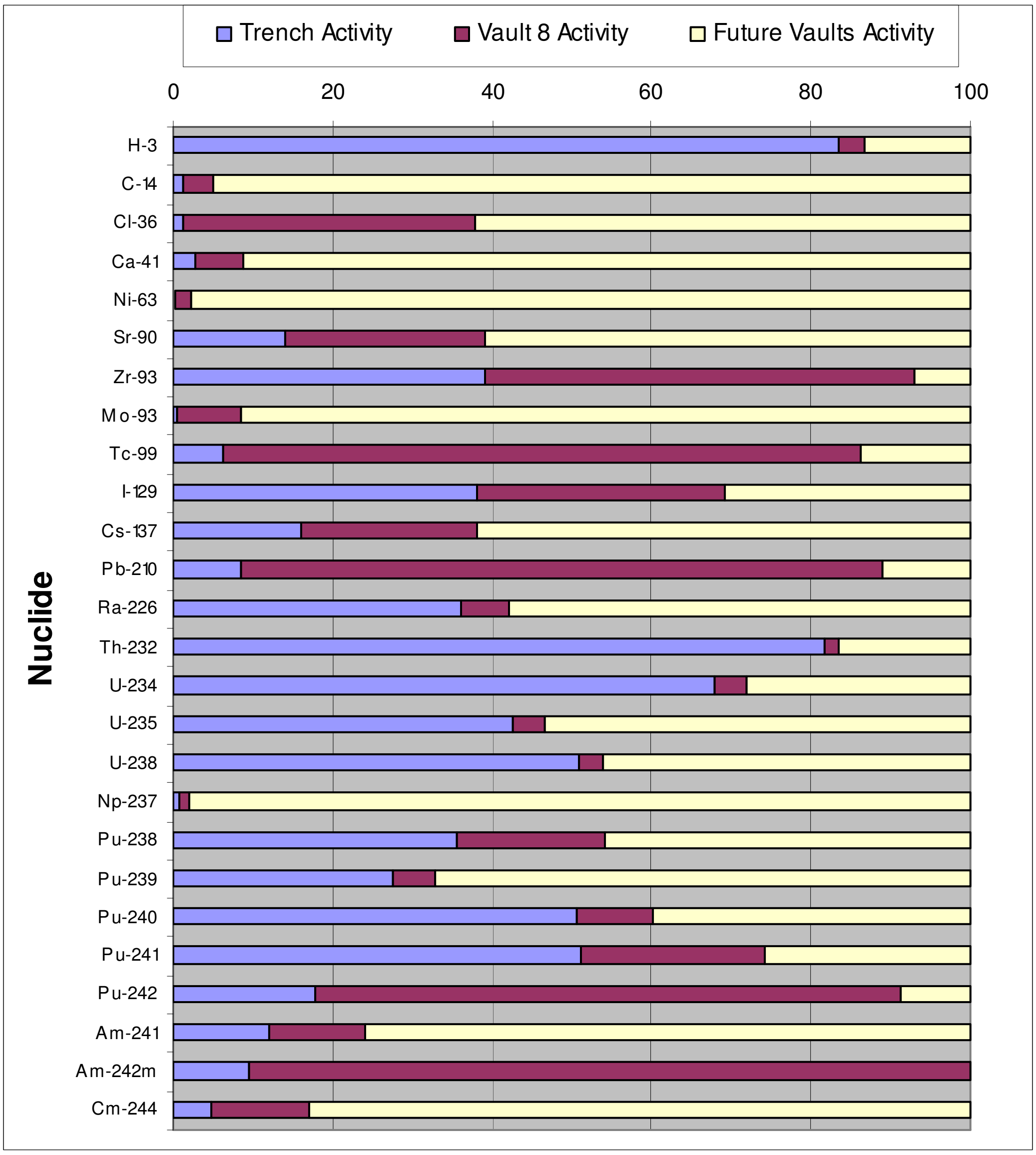

Figure 4.2 Comparison of Trench and Vault Activity Contributions (Wareing *et al.* 2008)

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Table 4.1 Changes in trench and vault activities for selected radionuclides (Wareing *et al.* 2008).

Nuclide	Activity Change Factor			
	Trenches	Vault 8	Future Vaults	Total
H-3	1.1	1.7	3.7	1.2
C-14	0.3	0.6	0.6	0.6
Cl-36	0.1	1.7	3.3	2.0
Ca-41	477	3.03	10.4	9.3
Ni-63	0.7	0.08	1.1	0.9
Sr-90	0.4	1.2	2.6	1.2
Zr-93	306	31.3	5.5	32.0
Mo-93	-	0.0045	0.0003	0.0003
Tc-99	0.07	5.0	0.5	0.8
I-129	10.7	1.8	11.3	4.3
Cs-137	0.5	1.3	2.8	1.3
Pb-210	0.003	0.07	0.01	0.02
Ra-226	0.5	0.22	1.7	0.7
Th-232	0.9	0.6	6	1
U-234	0.5	0.7	10	1
U-235	0.5	1.5	50	1
U-238	0.6	0.9	39	1
Np-237	0.8	2.0	303	53
Pu-238	0.3	0.6	1.5	0.5
Pu-239	0.9	0.8	11.6	2.3
Pu-240	0.7	0.7	3.4	1.0
Pu-241	0.4	0.6	0.9	0.5
Pu-242	0.8	1.36	1	1
Am-241	0.5	1.44	13	3
Am-242m	119	148	0.3	135.6
Cm-244	0.2	0.8	23	3
Total (all rads)	0.8	0.5	1.5	1.0

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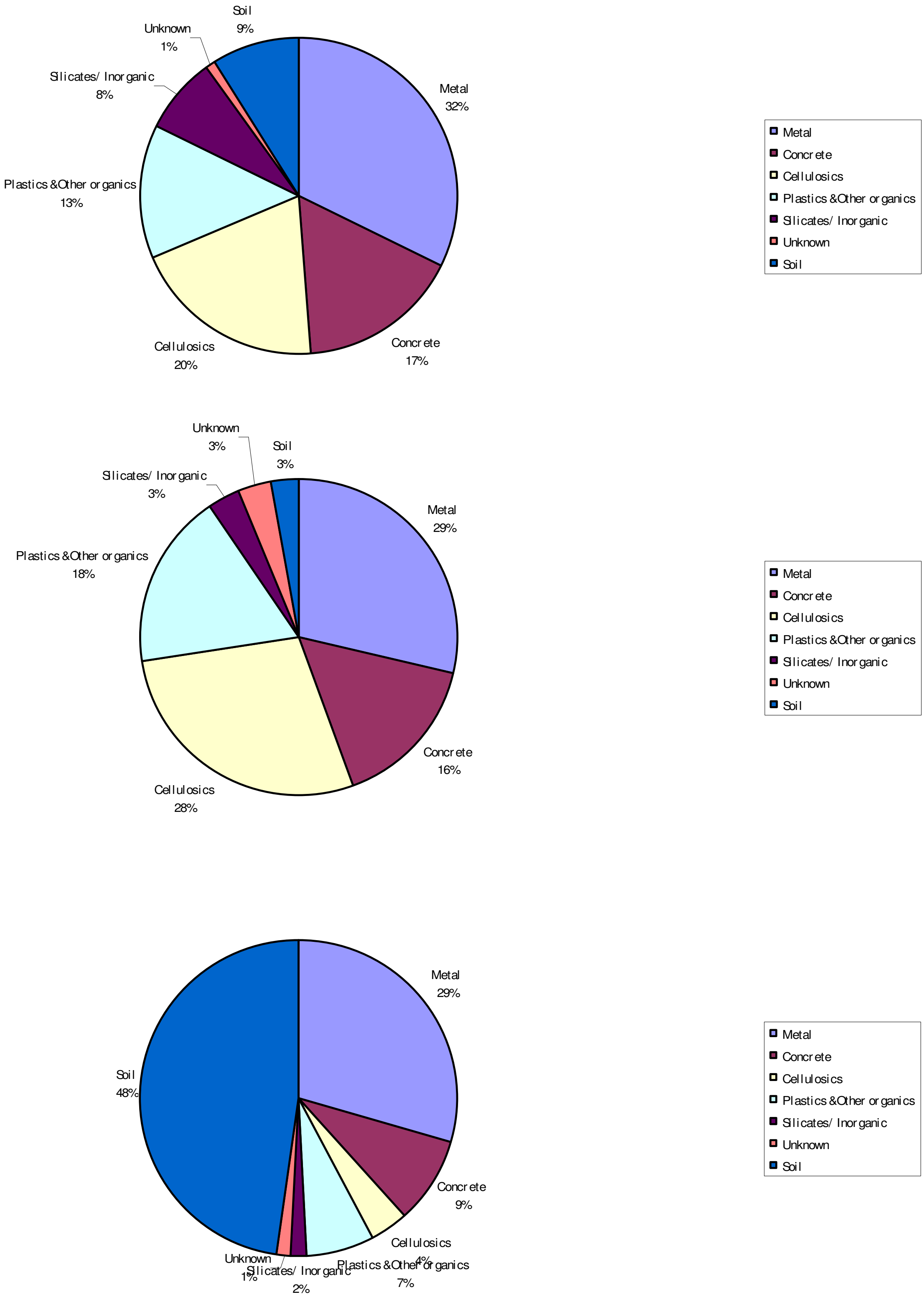

Figure 4.3 Distribution by volume of waste material in the trenches (top), Vault 8 (middle) and future vaults (bottom)

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4.4 Specific Radionuclides

4.4.1 Heterogeneity

The high level of discretisation present in the inventory has allowed a more detailed study of heterogeneity to be undertaken across the disposal system, including the identification of the location of specific consignments to the trenches. Clearly, this provides important information when discussing such issues as engineering optimisation, human intrusion or site end points.

Having data at such a high level of detail has allowed the construction of a number of schematic trench 'maps' for radionuclides and associated materials, to identify heterogeneity in distribution across the disposal system. These maps show the distribution of radionuclides on a bay-by-bay basis and are reported in full in Section 3.2 of Lennon *et al.* (2008). This information has been used to inform the assessments of the potential for targeted interventions of parts of the trench inventory, as reported in Volume 2, and also the detailed trench-scale geochemical modelling presented in Section 5 of this document.

As reported in Lennon *et al.* (2008), the majority of radionuclides are distributed homogeneously across the trenches. A summary of the heterogeneity within the trenches is shown in Table 4.2 – which can be read in conjunction with Figure 4.1, which shows the relative positions of the trenches.

From this table it can be seen that a number of radionuclides exhibit some level of variation across the trench system. Some of these, such as Nb-93m and Zr-93 are present in low activities. Of the radionuclides present at higher activities, it is clear that H-3, Th-232, Ra-226 and to a lesser extent uranium and plutonium isotopes also show some variations across the trenches. Disposals of these radionuclides have been associated with specific consignments whose locations have been identified.

Further discussions on individual radionuclides are provided below:

4.4.2 Uranium Disposals

The 2002 Safety Cases, in particular the 2002 PCSC, showed that uranium isotopes were key radionuclides determining the peak risks associated with the LLW disposals. For certain scenarios, the assessed impacts were higher than the risk target of 1 in a million per year. For the groundwater and gas pathways, uranium contributes to risks higher than 1E-6 y-1 with the remaining radionuclides orders of magnitude lower than this. Uranium isotopes were also important in all other exposure pathways. Consequently, uranium disposals to the LLWR were of primary focus for the update of the inventory and subsequent modelling activities.

Table 4.1 shows that, whilst the overall activity of uranium at the site remains the same as in 2002, the inventory of all uranium isotopes in the trenches has now reduced by a factor of two. This reduction has been due to the improved methodology of assessing the historical disposals to the trenches, in particular the capture of key consignment data from the disposal records. Of this uranium, disposals from the Springfields site represent about 95% of the total

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uranium trench inventory (Lennon, 2007a, b). The nature of this waste and the impact on the near field behaviour of uranium is discussed further in Section 5.

Table 4.2 Percentage breakdown of radionuclide inventory in the LLWR trenches.

Note: table is colour coded as follows: >14%, > 50%

Radionuclides	Trench 1	Trench 2	Trench 3	Trench 4	Trench 5	Trench 6	Trench 7
H-3	0.0	0.0	6.5	3.3	1.2	**89.0**	0.1
C-14	5.9	12.3	**20.4**	13.6	**15.4**	**15.9**	**16.4**
Cl-36	0.4	1.9	8.5	**15.0**	**20.4**	**30.3**	**23.5**
Ca-41	4.4	5.3	**19.0**	**18.0**	**23.4**	**18.6**	11.3
Ni-63	6.6	**15.1**	**18.0**	9.4	6.3	12.7	**31.9**
Sr-90	5.7	13.2	**15.3**	9.3	5.4	11.0	**40.2**
Zr-93	0.0	0.1	0.1	0.0	0.0	1.4	**98.4**
Nb-93m	0.0	0.0	0.0	0.0	0.0	0.6	**99.4**
Mo-93	4.6	5.6	**19.5**	**18.5**	**24.1**	**19.1**	8.6
Tc-99	**16.5**	14.0	12.4	5.8	2.9	7.7	**40.6**
I-129	7.7	**17.2**	**19.4**	8.3	5.2	12.1	**30.2**
Cs-137	4.9	11.3	13.5	**15.3**	6.7	9.6	**38.7**
Pb-210	5.2	**20.2**	**28.6**	11.5	13.8	13.1	7.5
Ra-226	0.4	**19.7**	**75.9**	1.1	1.3	1.1	0.7
Th-229	8.2	**18.5**	**20.3**	8.2	4.6	12.2	**27.9**
Th-230	7.7	**26.4**	**18.1**	9.7	7.5	11.7	**18.9**
Th-232	0.0	**36.8**	2.0	**32.0**	**28.6**	0.5	0.0
Pa-231	**16.4**	**20.8**	6.4	4.2	2.6	7.1	**42.4**
U-233	1.3	2.4	3.9	4.2	5.4	8.6	**74.3**
U-234	6.4	12.9	3.5	**34.6**	**26.0**	8.9	7.6
U-235	6.5	13.0	3.6	**34.3**	**25.9**	9.0	7.8
U-236	5.7	12.3	13.7	**29.9**	9.4	9.7	**19.2**
U-238	6.3	13.4	7.3	**32.0**	**24.0**	8.7	8.3
Np-237	**14.3**	**16.4**	8.8	4.4	2.4	7.5	**46.2**
Pu-238	2.3	**20.7**	6.7	**37.9**	10.9	6.3	**15.1**
Pu-239	1.8	**66.5**	4.7	11.8	3.8	3.6	7.8
Pu-240	2.0	**42.7**	5.5	**27.9**	8.1	5.0	8.8
Pu-241	1.9	**76.5**	4.8	2.4	1.4	3.1	10.1
Pu-242	5.6	**24.6**	12.5	9.4	7.6	13.8	**26.5**
Am-241	3.8	10.1	10.8	**34.6**	11.2	9.4	**20.1**
Am-242m	3.9	**25.2**	12.2	9.9	9.8	**16.4**	**22.6**
Cm-242	1.8	**15.9**	10.0	4.4	1.2	3.4	**63.2**
Cm-244	5.2	**16.6**	**16.2**	7.3	3.9	9.7	**41.1**

The dominance of one particular waste stream means that the distribution of uranium within the trenches is particularly heterogeneous, with Trenches 4 and 5 containing the majority of the uranium (Section 3.2, Lennon *et al.*, 2008). Additionally, 70% of the Springfields disposed uranium activity is located within just 91 bays of these trenches.

A second feature of the uranium inventory is that future disposals are anticipated to be significantly higher than was believed in 2002. This is due to the changes to the uranium activity declared for the Springfields decommissioning waste stream 2E101, which contributes just over 90% of the future vaults uranium inventory (Table 17 of Wareing *et al.* 2008). The result is that uranium in the future vaults inventory is now approximately equal to that in the

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trenches, whereas within the inventory calculated for the 2002 Safety Cases, uranium in the trenches accounted for around 90% of the total site inventory.

4.4.3 Neptunium Disposals

The disposal inventory of Np-237 is calculated to be significantly higher than that derived for the 2002 Safety Cases (Table 4.2). As shown in Figure 4.1, approximately 99% of this inventory is to be found in the future vaults inventory. This radionuclide has not been declared to any significant degree in the disposal records for waste to the trenches and Vault 8, whereas it is declared in the waste stream fingerprints for future arisings in the 2004 National Inventory, in particular for the 2E101 waste stream representing decommissioning LLW from the Springfields site, which contributes almost 100% of the Np-237 inventory in the future vaults (Table 20 of Wareing *et al.,* 2008). This waste stream also contributes over 90% of the future uranium disposals of uranium and so represents the main contributor of potentially significant alpha emitting radionuclides to the future disposal inventory.

4.4.4 Thorium Disposals

The majority of the thorium activity in the derived LLWR inventory is located in the trenches and is due to specific disposals of mineral sands and process residues (Section 3.2, Wareing *et al*., 2008). This includes thorite sands disposed during 1972 and 1973, monazite sand in 1960 and thorium-contaminated waste disposed during specific periods (1965 to 1967, 1973 to 1977 and 1979 to 1983). Consequently, there is some degree of heterogeneity associated with thorium disposals in the trenches (Section 3.2, Lennon *et al.,* 2008) with thorium concentrations localised within a small number of bays. For example, Lennon *et al*. (2008) show that whilst activity is distributed fairly evenly across Trenches 4 and 5, there are a small number of bays in Trench 2 that contain between 1 and 10% of the total Th- 232 activity in the LLWR trenches. These are due to the disposal of mineral ores with a high thorium content. The thorium-contaminated waste also contained the majority of the radium disposals to the trenches. In contrast, Th-232 activity in the vaults is expected to be significantly lower.

4.4.5 Carbon-14 Disposals

Approximately 98% of C-14 at the LLWR will be present in the future vaults (Figure 4.2). C-14 is an activation product present in graphite, steel and concrete arising from the decommissioning of nuclear reactors, with the majority of this waste not generated until after 2040. It should be noted that the total predicted activity of 7.5 TBq for C-14 would not be acceptable under current disposal limits, and it is possible that alternative solutions will be found for its disposal. However, this activity has been included in full within the inventory to allow an assessment of its impact.

4.4.6 Radium Disposals

The derived Ra-226 activity is distributed approximately equally between the trenches and the vaults. The majority of radium activity in the trenches is due to specific disposals of process residues (namely the thorium-contaminated waste described in Section 4.4.4) whereas activity in the vaults will arise mainly from waste generated during land remediation. The distribution of Ra-226 within the trenches is extremely heterogeneous, with 95% of the total activity present in Trenches 2 and 3. The distribution of activity shown in Lennon *et al.* (2008) also shows there to be a small number of bays containing between 1 and 10% of the total Ra-226

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activity, with one bay within Trench 1 containing over 10% of the total Ra-226 activity in the trenches.

4.4.7 Tritium Disposals

Over 80% of the LLWR tritium inventory is found within the trenches. Of this, the disposal of Betalights between December 1983 and April 1984 to Trench 6 accounts for the majority of tritium in the trenches.

4.4.8 Plutonium Disposals

Plutonium is a minor contaminant of a range of operational wastes. Within the trenches, Pu-239 and Pu-241, in particular, are disposed mainly within Trench 2, with the remaining plutonium evenly distributed across the remaining disposal areas. Almost 90% of the future vaults Pu-239 inventory is from contaminated land from Aldermaston, which is also the majority contributor of Ra-226/Th-232 activity (Table 21 of Wareing *et al*., 2008).

4.4.9 Other Radionuclides

The inventory for a small number of radionuclides has changed by several orders of magnitude; namely Ca-41, Zr-93, I-129 and Am-242m, which have all increased in the trench inventory, whilst Am-242m has increased in the vaults inventory.

4.5 Summary

The inventory of disposals at the LLWR has been re-evaluated as part of the current Lifetime Project. A key feature of this re-evaluation has been a widening of the range of sources used to extract disposal data, particularly for the historical inventory.

The resulting inventory represents a significant improvement in knowledge of the nature of disposals at the LLWR site. In particular, the historical trenches inventory is much more robust. The trench inventory was calculated for the 2002 Safety Cases largely by backfitting radionuclide and materials inventory data declared in the National Inventory for contemporary waste streams to known historical consignor volumes. Within the current work, actual disposal records have been primarily used to develop the trench inventory. The new methodology has allowed precise mapping of the disposals inventory to locations within the trenches, both by radionuclide activity and material contents, with a consequent understanding gained of the heterogeneity of waste.

Application of this new methodology has resulted in a reduction in the overall trench radionuclide inventory by around 20% from that calculated for the 2002 Safety Cases. Significantly, the trench uranium has now reduced by a factor of two, with the vast majority of this uranium inventory associated with disposals from the Springfields site. This is anticipated to reduce risks calculated for uranium in the radiological safety assessment, which were shown to be of key significance in the 2002 Safety Cases.

The inventory for Vault 8 has been calculated almost entirely from actual disposal data, with minimal support provided by the detailed radionuclide and material fingerprints from the 2004 UK National Inventory where necessary. The Vault 8 inventory is available by individual

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consignment and therefore the data to underpin a detailed waste heterogeneity study is available.

The methodology for the calculation of the future vaults inventory has been improved from the 2002 Safety Cases to take account of operational data available in the Waste Tracking System database; in particular to influence the packaging model. An examination of the waste streams contributing the majority of activity in each of the radionuclide groups for the future vaults inventory has shown that between 70% and 90% of activity is represented by just five key waste streams, several of which are common to more than one group. In particular, just one Springfields decommissioning waste stream with a volume of approximately 3,000 m^3 contributes almost 90% of future uranium activity and almost all future Np-237 activity. It is important to note that the derivation of the future vaults inventory has been based on the waste streams given in the 2004 National Inventory where the future disposal route is declared as the LLWR. Therefore, no assumptions have been made regarding any changes to future conditions for acceptance or national LLW management strategy. However, the identification of significant future waste streams and waste types, such as the large quantities of contaminated soil scheduled for disposal at the LLWR, provides important information that could be used by the LLWR when considering the future use of the remaining disposal capacity of the site (see Volume 1 and 2).

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5 Near Field Behaviour

5.1 Introduction

An understanding of the evolution of the near field system is important as it controls, to a large extent, the behaviour and release of radionuclides within the disposed inventory. Radionuclides such as uranium, technetium and plutonium can exist in several oxidation states and exhibit different behaviour under the range of redox potentials and pH conditions that may occur in the near field environment.

A detailed understanding of the biogeochemistry of the trench and vault systems was established as part of the 2002 PCSC. This work is presented in BNFL (2002c), which describes the evolution of the near field as a result of a complex set of interactions between waste degradation, metal corrosion, cement degradation, microbial reactions and groundwater. The representation of the near field evolution was provided primarily through the application of the biogeochemical model DRINK, which has the capability of simulating all the processes described above. However key support, in terms of developing the conceptual model and provision of numerical parameters for the model, was provided by an extensive set of experimental and underpinning modelling studies.

As highlighted in Section 3, changes to our understanding of the climate and landscape evolution may have some influence on the evolution of the site. Additionally, the recent work evaluating the performance of the engineering features of the site presented in Section 6 indicates that the trenches in particular will be at least partially saturated. The 2002 DRINK model had assumed a greater degree of saturation and so again some modification of our understanding of the biogeochemical evolution of the site may be required before the submission of the full 2011 Environmental Safety Case. Nevertheless, much of the understanding gained within the 2002 PCSC remains relevant for the current LLWR Lifetime Project. In particular, the main features of the estimated evolution of the trenches and vaults have been assumed to be valid for the current assessment. However, significant progress has been made since 2002 in further understanding and quantifying the evolution of the near field system, and in particular the behaviour of key radionuclides within the disposed inventory. Much of this increase in understanding has been made possible through the increased knowledge of the activity and nature of the disposed inventory described in Section 4.

The following sections provide an overview of the progress that has been made during the Lifetime Project.

5.2 Trench biogeochemistry

Before presenting the progress that has been made since 2002 in our understanding of the near field environment, it is useful to summarise the main points that arose during the 2002 PCSC. The 2002 PCSC near field biogeochemistry report (BNFL, 2002c) provides a detailed description of this understanding of the biogeochemical processes operating within the LLWR trenches and the numerical model used to represent these processes. This understanding is summarised in Section 3 and can be generalised as:

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- Establishment of reducing and acidic conditions due to degradation of cellulosic and metallic waste material. This period is characterised by the production of reduced chemical species such as methane, acetate and sulphide.

- By approximately 4000 years, the trenches re-oxidise to be consistent with the conditions encountered in the geosphere.

The main radionuclides affected by the development of these biogeochemical conditions are those which have mixed valency and are thus sensitive to changes to the redox conditions within the near field. For the 2002 PCSC the key radionuclides that exhibited this behaviour included neptunium, technetium and, particularly, uranium.

The results of the 2002 PCSC revealed that the greatest impact presented by the disposed inventory was associated with uranium isotopes, particularly for the groundwater pathway and for human intrusion, with other radionuclides such as Th-232 making significant contributions to the coastal erosion and human intrusion pathways. The risks from U-234 and U-238 within the trench inventory frequently exceeded the risk target of 1 in a million per year for the groundwater pathway, in many cases being significantly above this target (reaching values of 10^{-4} y^{-1}). Consequently, a major focus of effort since 2002 has been on gaining further understanding of the nature of the uranium waste disposed within the trenches at the LLWR site.

Within the 2002 PCSC, the near field was represented through the DRINK biogeochemical model (BNFL, 2002c). Within this representation, all radionuclides, in the absence of detailed information on the physical and chemical nature of the wastes, were modelled conservatively in that they were allowed to initially dissolve in groundwater and subsequently precipitate as secondary mineral phases if prevailing geochemical conditions allowed. There was, therefore, no representation of the nature of the disposed waste form, which would be expected to limit the rate of dissolution into groundwater.

In the case of uranium, the DRINK model simulated that under the prevailing reducing conditions developed within the trenches, uranium release was controlled by the solubility of the U(IV) mineral phase UO_2 at concentrations ranging from 1E-4 to 1E-8 mol l^{-1}. These conditions were estimated to last for up to 4000 years, after which more oxidising conditions occurred and uranium was present in the more soluble U(VI) oxidation state. This resulted in a much higher flux of uranium from the trenches, leading to high risks above 1E-6 y^{-1} for the groundwater pathway.

Sensitivity analyses aimed at identifying the key processes controlling these risks (BNFL, 2002d) revealed that the peak risks for the groundwater pathway were crucially dependent on uranium behaviour. For example, a fixed uranium solubility of 1E-7 mol l^{-1} resulted in a risk of 1.6E-7, which is 1.7% that of the best estimate case. Similarly, restricting the amount of the uranium that was available for release also reduced the peak risk. On the basis of these studies it can be concluded that, within the 2002 PCSC, the groundwater risk would have met the 1E-6 y^{-1} target if:

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- Uranium concentration in the leachate was below 1E-6 mol l^{-1};

- The inventory of uranium available to dissolve was about 10% of that considered in the 2002 calculations; or

- The total flux of U-234 was around 1E-3 mol y^{-1}.

As discussed in Section 4, the work on the inventory within the LLWR Lifetime Project has shown that the trench uranium inventory has reduced by approximately 50%, due to more reliance on actual disposal records and less use of backfitting calculations. Additionally, the uranium is predominantly comprised of disposals from the Springfields site, which make up 95% of the trench inventory. In order to develop a more realistic model of the release of uranium and the long term behaviour of uranium within the trenches, an examination of the nature of this waste has been undertaken (Small *et al.,* 2008a).

Through an examination of literature dating back to the 1950s, it has been possible to develop an understanding of the waste processing practices undertaken at the Springfields site and therefore the nature of the uranium waste disposed within the LLWR trenches. This has revealed (Section 4, Small *et al.,* 2008a) that the majority of the Springfields disposals are made up of process residues including:

- Filter cake residues of the yellowcake and ore refining process, characterised as insoluble silicate material: and

- Magnesium and calcium fluoride residues resulting from the reduction of UF_4 to uranium metal.

These residues were subject to an aggressive treatment process, involving leaching in hot concentrated nitric acid, to recover as much uranium before disposal. This treatment process therefore removed the majority of the leachable uranium from the residues, leaving behind only small quantities (around 0.1 to 0.9 wt%) of highly insoluble uranium bound within a fluoride matrix. Therefore, the majority of the uranium disposed within the LLWR trenches is in a highly insoluble waste form.

To examine the behaviour of this waste form within the LLWR trenches, a conceptual and numerical model has been developed (Section 5, Small *et al.*, 2008a). This conceptual model is presented in Figure 5.1, with the key features being:

- Uranium is present mainly as metallic inclusions (or “shots”), which represent fine grained uranium metal retained in the reduction slag.

- A smaller proportion of uranium may be present dissolved as a solid solution in the fluoride matrix.

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- Release of the uranium occurs through the dissolution of the surface of the fluoride matrix which releases the uranium present as a solid-solution within the matrix and exposes uranium shot and other inclusions to groundwater.

A kinetic dissolution model to represent these processes has been implemented within the geochemical code PHREEQC (Parkhurst and Appelo, 1999). Model calculations indicate that the dissolution of MgF_2 residue containing 0.1 wt% uranium will lead to a dissolved uranium concentration of around 1E-7 mol l^{-1} in leachate. This is lower than the solubility of uranium estimated for much of the 2002 PCSC results, and is lower than the concentrations estimated to result in risks below the 1E-6 y^{-1} risk target.

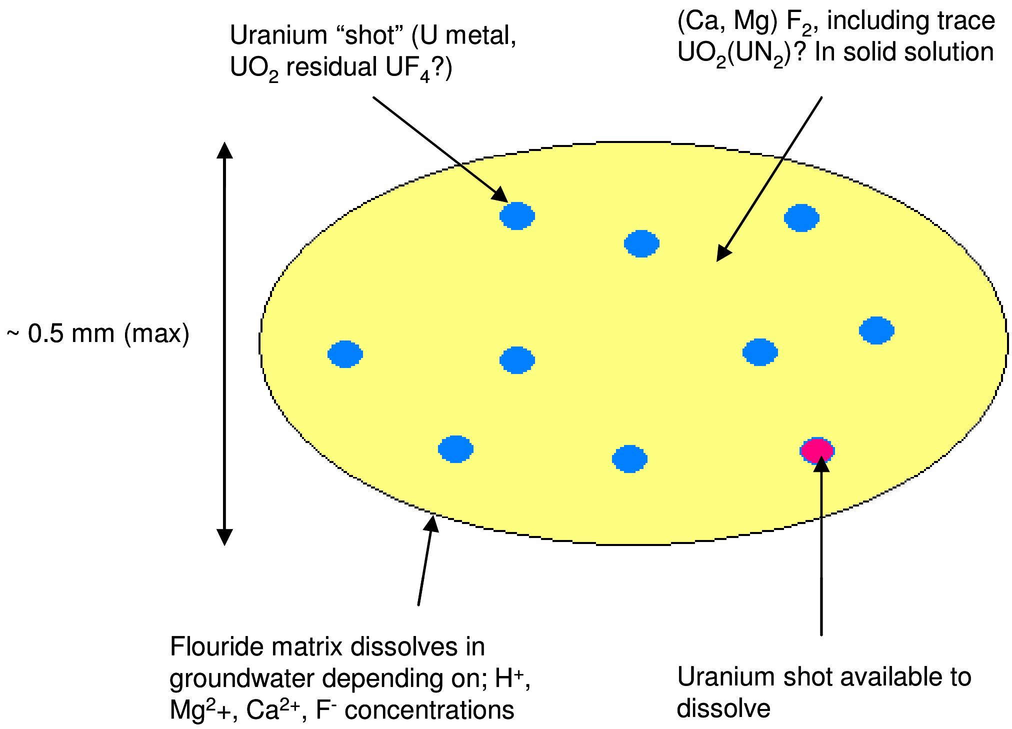

Figure 5.1 Schematic representation of the fluoride residue material.

In representing the behaviour of uranium release through this conceptual model there are clearly assumptions that have to be made. In particular, it is assumed that the slag is composed of pure MgF_2, whereas it is possible that other metals, such as iron and zinc, may also be present. There is evidence (Section 6.2 of Small *et* al., 2008a) that the presence of these metals may increase the solubility of the fluoride. Additionally, it has been assumed that fractures in the fluoride grains (which could lead to uranium being more readily leachable) have been assumed to be not significant. The supporting data presented below would indicate

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that these assumptions are reasonable. Nevertheless, further work could readily be done to further justify these assumptions, should uranium leaching behaviour prove important for the groundwater pathway.

However, the model has been given convincing support through consideration of site monitoring data, including analyses of trench leachate, trench vent probe samples and samples collected in geosphere boreholes located close to the LLWR trenches. Trench leachate samples contain significant concentrations of fluoride up to 30 mg l^{-1}, which is above that of natural groundwater, indicating that there is a source of fluoride from within the trenches. Geochemical interpretation of this data (Section 6, Small *et al.*, 2008a) indicates that the leachates approach chemical equilibrium with CaF_2 and are consistent with the dissolution of MgF_2 (i.e. the leachate data define a trend of increasing magnesium and fluoride activity in the mole ratio of 1:2) (see Figure 5.2).

Therefore, the reaction is defined by the release of magnesium and fluoride through the stoichiometrical dissolution of MgF_2, and the subsequent reaction of fluoride with calcium in groundwater to precipitate CaF_2, as represented by the equation:

$$Ca^{2+} + MgF_2 = CaF_2 + Mg^{2+}$$

Uranium groundwater concentrations are also frequently correlated with fluoride, in particular within regions at the northern end of trenches 1, 4, 5 and 6, which provides further support for the conceptual model. In particular, the highest uranium concentration is associated with a sample from a trench vent probe within Trench 6 (Vent probe 6.3), which also has the highest fluoride concentration (see Figure 5.2). These fluoride concentrations are above the solubility of both magnesium and calcium fluoride. High concentrations of zinc are recorded in leachates and it is known that zinc fluorides are more soluble than magnesium fluorides. Therefore, zinc (or other impurities) could conceivably be present, increasing the solubility of the fluoride and thus the aqueous concentration of uranium.

In contrast, within Trench 7 high uranium concentrations are not associated with high fluoride concentrations. This indicates that some other uranium waste form is present in this region and is consistent with the discussion within Lennon (2007a) (and summarised in Section 4 of this report), which shows that the Springfields disposals are predominately found within Trenches 4 and 5. These may be associated with disposals of the more soluble UF_6, for example.

Significantly, fluoride is also detected in high concentrations in geosphere boreholes close to the trenches. In particular, the cluster boreholes C1 and C2 (for the location of these boreholes see Volume 4) are notable for fluoride concentrations above that of the local groundwater. These boreholes are also associated with high tritium concentrations (see Volume 4) and indicate that fluoride, as a conservative tracer within groundwater, has leached from the trenches into the geosphere, providing a tracer of the hydrogeological pathway from the uranium fluoride residue disposals.

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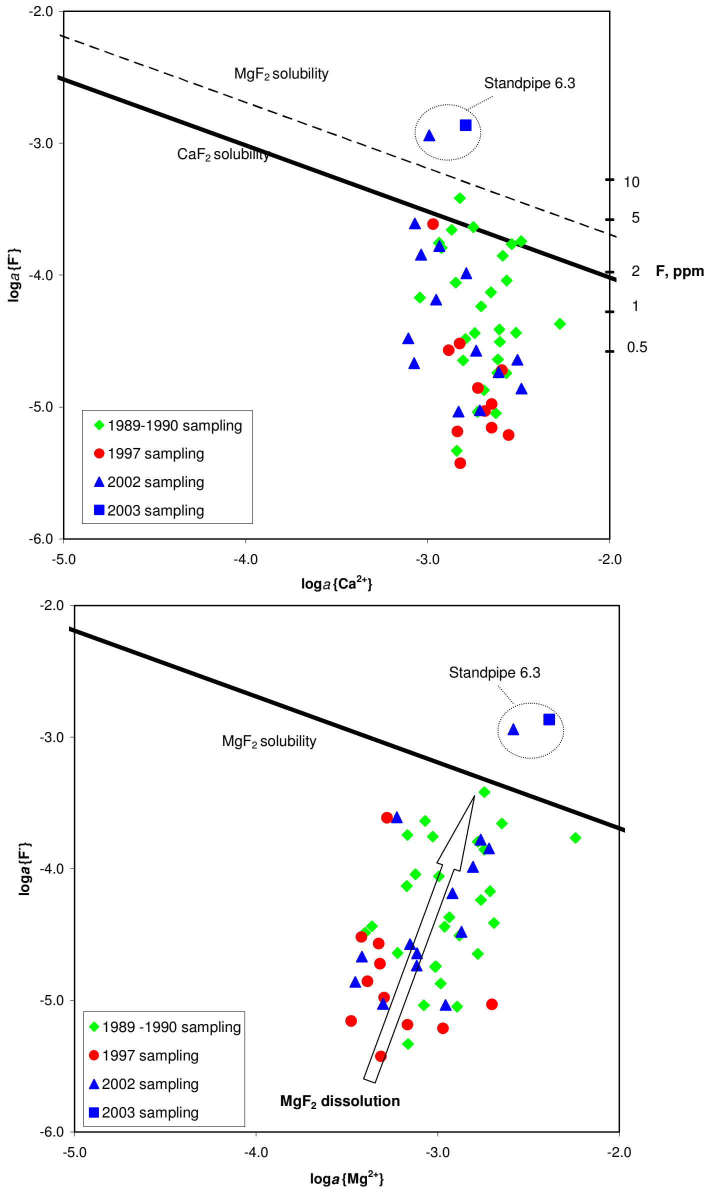

Figure 5.2 Cross Plots of Mg (top) and Ca (bottom) ion activities against fluoride ion activity from the analysis of trench leachate and comparison to the solubility of MgF_2 and CaF_2.

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Detailed modelling of the trenches environment has been undertaken to examine the behaviour of these fluoride residues, incorporating local hydrogeological effects and features within each trench, including the location of firebreaks, soil between the trenches and specific locations of uranium disposal (Small *et al*., 2008a; Kwong *et al.,*2008; Kwong, 2008). The results of the models, using the PHAST computer code (Parkhurst *et al*., 2004), indicate uranium concentrations consistent with the measurements of trench leachates. For example, Figure 5.3 compares the frequency distributions of the concentrations of uranium measured in trench leachate with the results of one such model of uranium dissolution (Small *et al*., 2008a). The model distribution reflects regions where the solid fluoride is present, which have highest uranium concentrations ($10^{-6.4}$ mol kgw^{-1}), and the downstream plume in the trench zone ($10^{-7.9}$ mol kgw^{-1}). These two populations overlap the concentration range of the site data set. Two samples with highest measured uranium concentration above $10^{-5.5}$ mol kgw^{-1} are from Trench 6 (Vent probe 6.3) which may be associated with other more soluble types of fluoride wastes. The latest of these models (Kwong, 2008) has also successfully integrated the uranium dissolution representation with a realistic groundwater flow model, parameterised to measured groundwater levels within the trenches and the wider vicinity. It is anticipated that near field models such as these will provide vital information in support of the full Environmental Safety Case scheduled for 2011.

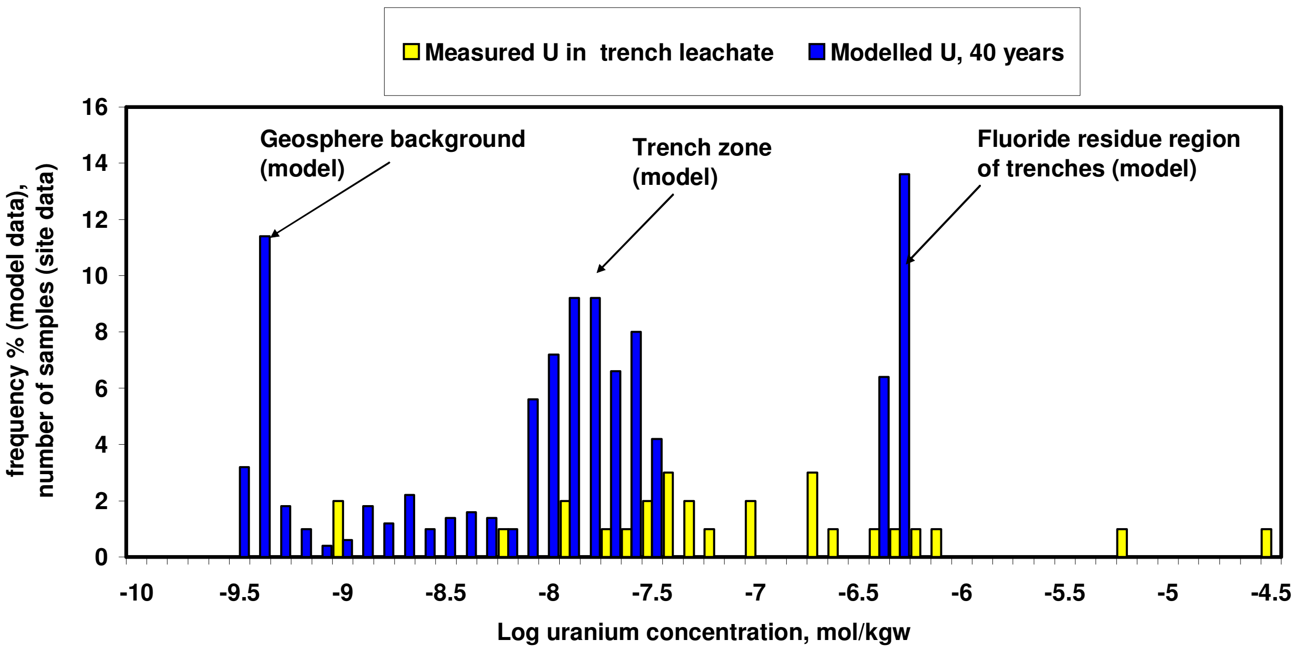

Figure 5.3 Comparison of modelled uranium concentrations and leachate concentrations

The effects of variations in waste heterogeneity on the biogeochemistry and uranium solubility in the LLWR trenches have also been examined (Kwong *et al.,*2008) through use of a finer scale grid than was used for the 2002 PCSC calculations, again with explicit representation of

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firebreaks and soil regions between the trenches. This representation indicated that small scale heterogeneities within the trenches, producing, for example, adjacent areas of oxidising and reducing environments, will result in mobilisation and re-precipitation of uranium. These "uranium roll fronts" are frequently observed in natural systems and will slow the release of uranium during the period when the trenches re-oxidise. It can therefore be concluded that the 2002 biogeochemical model, which considered the system at a significantly larger scale, overestimated the release of uranium.

The updated information on uranium release from the trenches, limited by the dissolution of fluoride waste residues, will have a significant impact on the flux of uranium from the near field. The current uranium concentrations in trench leachate are likely to be maintained at the levels described above and the groundwater uranium flux from the site may be lowered by the emplacement of a post closure cap and other engineering, which will reduce water flow through the site. The consequent flux of uranium from the trenches should therefore be lower than that estimated within the 2002 PCSC, particularly after the re-oxidation of the trenches, lowering the groundwater risk presented by the disposal of uranium.

5.3 Vault Biogeochemistry

The results presented within BNFL (2002c) demonstrate that the evolution of the vault system is expected to be dominated by the dissolution of the large amount of cementitious material, present predominantly as grout backfill, and the corrosion of metal, from the ISO freight containers and from directly disposed metal waste. This will result in the development of high pH and reducing conditions. The establishment of these conditions is supported by a range of experimental simulant studies (Trivedi and Dutton, 2008). The modelling results reported in BNFL (2002c) indicate that alkaline conditions would prevail for 10,000 years and would retain many of the key radionuclides through solubility and sorption processes. Uranium, for example, disposed in the vaults has a much lower radiological impact for the groundwater pathway compared to the trenches due to its lower inventory and solubility.

The model presented in BNFL (2002c) represented the vault system as being homogenous, with a large grid size, and did not explicitly model the interactions between the waste and the cement grout. To examine whether these assumptions are of major importance in representing the evolution of the vault system, a more recent modelling study has been undertaken as part of the LLWR Lifetime Project (Kwong *et al.* 2008).

The model considered a 2m square region of a vault ISO container containing 16 waste pucks enclosed in a cement grout backfill (see Figure 5.4). The cement grout was considered to buffer pH to alkaline conditions through the dissolution of calcium silicate hydrate (or CSH), which is leached by the inflowing groundwater to release calcium, which can precipitate as calcite ($CaCO_3$). Alkaline conditions are maintained as long as CSH remains within the grout and so determining the time for this mineral to completely dissolve was an important part of this modelling study.

The results demonstrated that some heterogeneity is observed within the system. The region of the model representing the grout backfill is characterised by a pH of 11, whereas the waste pucks have a pH of around 9, resulting from the infiltration of alkaline groundwater (Figure

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5.5). Uranium is shown to be largely soluble within the waste pucks but has a low concentration (1E-8 mol l^{-1}) in the more alkaline conditions within the cement grout (Figure 5.5), where it is limited by the precipitation of $CaUO_4$. This indicates that the uranium largely precipitates within the grout around the outside of the waste pucks.

CSH phases in the backfill grout are shown to progressively dissolve within the system, its dissolution accelerated by the CO_2 generated within the waste pucks. Although the CO_2 concentration is assumed conservatively to be constant over time, the model predicts that a total of 2600 years is required to dissolve the CSH phases from this 2m block of cemented waste form. In reality, it would be expected that the rate of production of CO_2 would decline over time, which would mean that the buffering effect of the CSH phases would persist for much longer.

These preliminary results support the conclusions of the 2002 PCSC that the vault system is characterised by high pH conditions. Although some heterogeneity will exist within an individual ISO container, the overall vault system will remain buffered to a high pH for many thousands of years and may be considered homogeneous for the purpose of assessment calculations. The backfill will provide an effective barrier to the release of radionuclides, through the precipitation of insoluble mineral phases and the high sorption potential presented by the surface of cementitious materials.

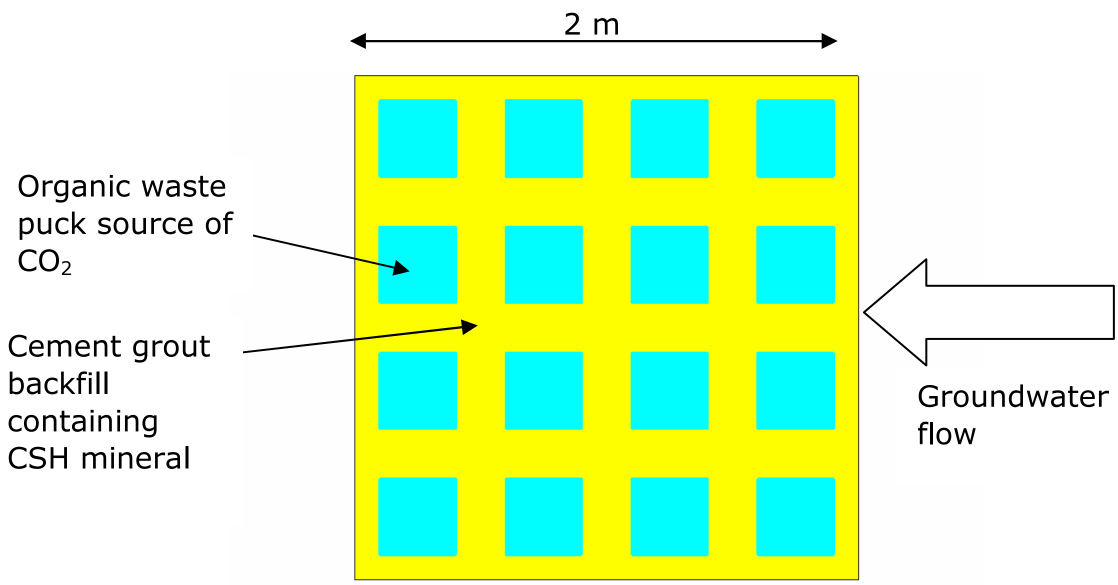

Figure 5.4 Schematic of the vault waste form model

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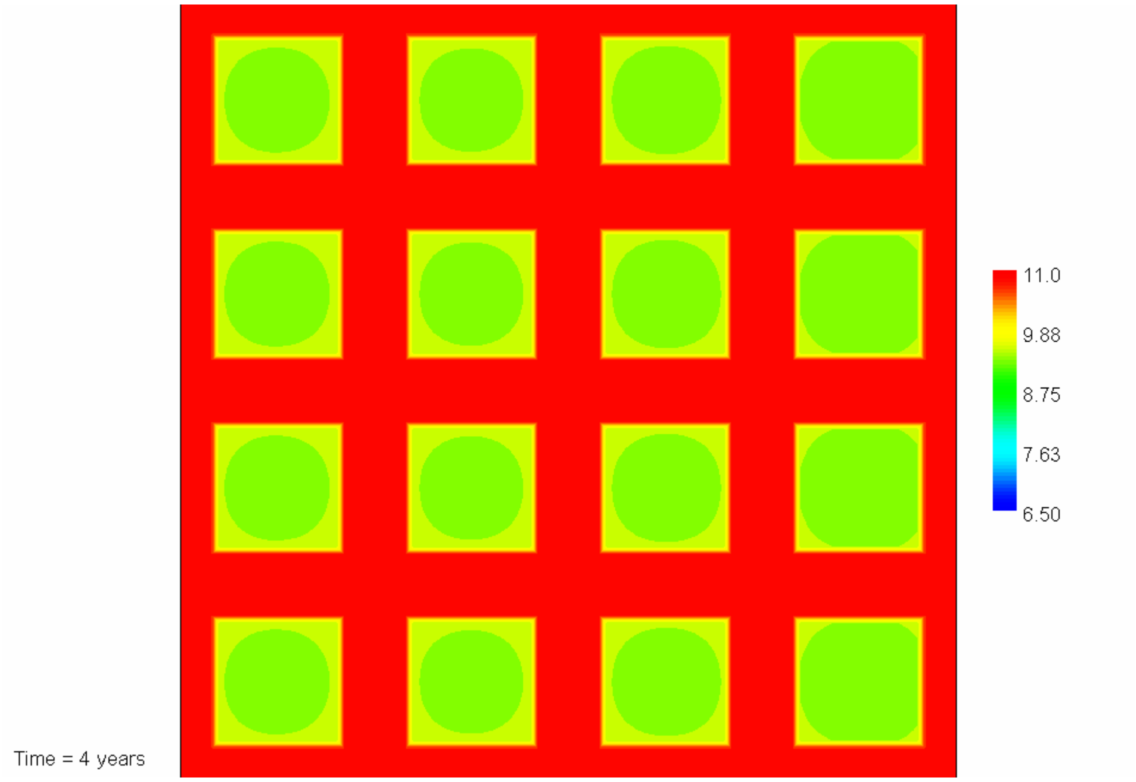

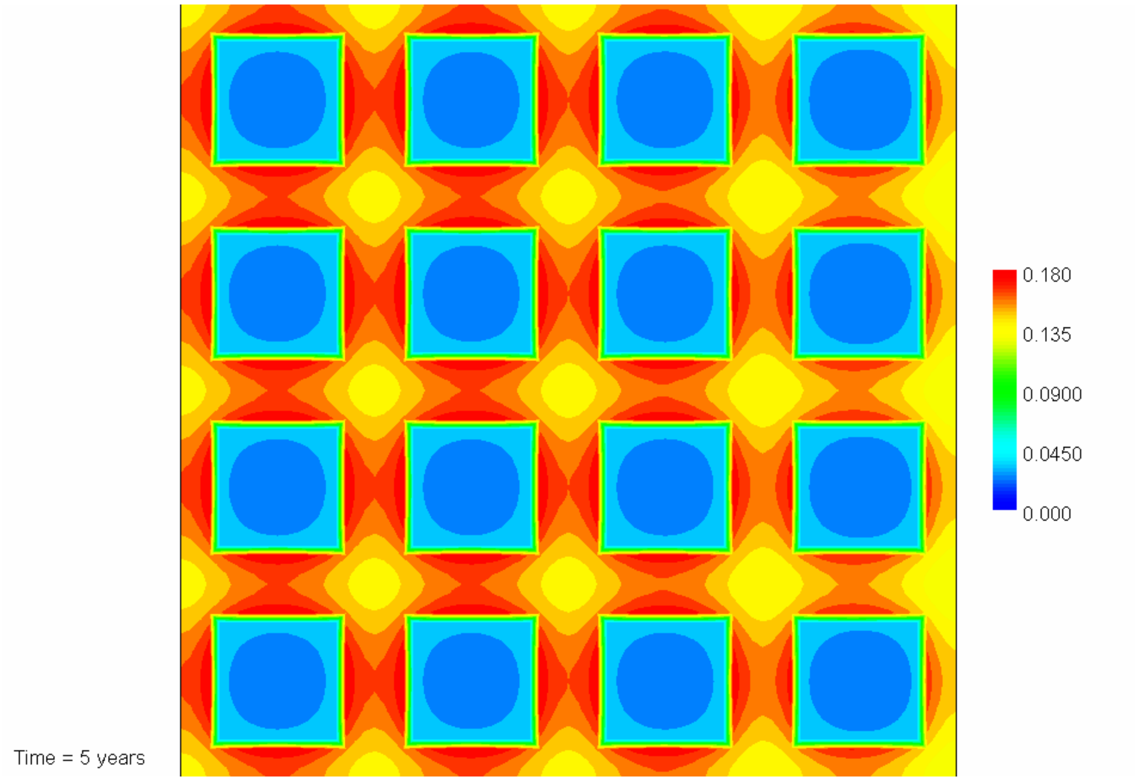

Figure 5.5 Calculated pH distribution amongst the waste pucks and cementitious grout (top) and modelled $CaUO_4$ precipitation (mol kgw^{-1}) (bottom)

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Additional work focussing on the vault system undertaken within the LLWR Lifetime Project has included a review of the potential influence of additives used in the cement grout of the vault waste form. These additives are included within the grout to decrease viscosity and thus ensure effective pore filling, minimising the potential for significant settlement and cracking of the final site closure cap.

The additive used within the LLWR grout formulation is Sikament 10, a vinyl co-polymer based superplasticiser. Some literature evidence exists that indicates that this, and other superplasticisers, have the potential to increase the mobility of radionuclides. The Environment Agency highlighted this issue during their review of the 2002 PCSC (Environment Agency, 2005), and included it within IAF NRF_012.

Consequently a comprehensive literature study (Trivedi *et al.*, 2008) has been undertaken to assess any impact of the use of superplasticiser. This revealed that some experimental studies have shown that the presence of superplasticiser has resulted in an increase in radionuclide mobility (e.g. Glaus and Van Loon, 2004; Boult *et al.*, 1998). However, these experiments were carried out with the superplasticers in solution and therefore represent a worst case scenario. Additionally, Dario (2004) undertook batch distribution experiments to assess the potential effects of a number of cement additives, including Sikament 10 on the mobility and distribution of radionuclides with the conclusion that relatively high concentrations of Sikament 10 are required to significantly reduce the sorption of Eu. Experiments in which the additives are incorporated within cured cement show no increase in radionuclide mobility, probably due to the slow diffusion rates in cement of these large organic molecules limiting their concentrations at the cement surface to negligible levels (e.g. Greenfield *et al.*, 2000). These diffusional effects are likely to be most significant in pozzolanic cements, such as the LLWR grout.

There is therefore potential for superplasticisers to influence the mobility of radionuclides within a cementitious environment. However, there is no clear view from the literature to draw any firm conclusions on whether there will be a significant effect on radionuclides within the LLWR disposal system. However, the majority of experimental studies that demonstrate some increase in radionuclide mobility used free solutions which significantly over-represents the additive concentration in cement/grout porewater. It is therefore concluded that the effect of superplasticiser in the LLWR grout enhancing the mobility of radionuclides is likely to be small.

5.4 Colloids and organic complexation

Colloids are solid materials with particle sizes between 1µm and 1nm in diameter that are suspended in aqueous solution. Within a disposal assessment context they could comprise radioelement particles/aggregates (so called “true colloids”), and inorganic and organic materials such as clays, metal oxides, organic macromolecules (humic and fulvic material) and suspended microbes, upon which radionuclides can sorb forming “pseudo-colloids”. Colloidal material has the potential to be transported at the velocity of groundwater flow without retardation. For this reason it is important to consider the possibility of colloidal transport in the groundwater at the LLWR.

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Field based investigations of the occurrence, physical characteristics and the association of radioactivity with colloidal material were undertaken between 1999 and 2002 (Warwick, 1999; Warwick 2000; Eilbeck and Warwick 2003a; Eilbeck and Warwick 2003b). The particle size, number and composition of colloidal material were investigated by photon correlation spectroscopy and Scanning Electron Microscopy (SEM) analysis for inorganic colloidal material and by spectroscopic techniques to characterise humic and fulvic material. The association of radioactivity with colloidal material was investigated by alpha and beta analysis and gamma spectroscopy. Results from this analysis clearly demonstrated the presence of colloidal material in both the near-field and geosphere environments. There were enhanced levels of both inorganic and organic colloids in the near field. A fraction of the radioactivity in the trenches, particularly alpha, was associated with filterable activity. However no radioactivity was associated with geosphere colloids although the samples were taken from boreholes associated with the highest levels of contamination from plumes originating in the trenches. Experimental work also suggested that near-field colloids may not be stable when migrating from highly anaerobic near-field conditions to a more aerobic geosphere environment. Assessments reported in 2002 (BNFL, 2002c) considered the effects of inorganic colloids to be less significant than other near-field uncertainties, such as solubility and explicit consideration of their effects in subsequent assessments may not be warranted.

The effects of natural humic and fulvic organic colloids were not evaluated to the same level of detail as inorganic colloids in the 2002 assessment. However, Trivedi *et al*. (2008) have examined the interaction of uranium with aqueous humic and fulvic species at concentrations measured at the LLWR. Humic and fulvic species are large organic macromolecules of variable size and composition, which are formed through the natural degradation of organic matter. Their potential effect can be evaluated in a simplified manner by geochemical speciation modelling. Trivedi *et al*. (2008) have modelled the ability of these species to complex to radionuclides. The modelling studies undertaken have shown that, while humate ions can form complexes making up 6% of total dissolved uranium, this is only under very specific combinations of conditions that are unlikely to occur. The modelling study indicated that fulvate ions do not form any significant complexes under any modelled conditions. On this basis, it has been shown that organic macromolecules are unlikely to have a significant effect on near field solubility and sorption of uranium, a key risk and dose determining radioelement in the LLWR trenches from the 2002 PCSC.

5.5 Summary

Recent work within the LLWR Lifetime Project has examined the assumptions associated with the representation of the near field system. This has included the nature and behaviour of the uranium waste disposed in the LLWR trenches, the representation of small scale heterogeneities within the trench environment and the interactions of waste and the grout backfill within the vaults.

The key conclusion from this work is that the uranium fluxes from the trenches calculated as part of the 2002 PCSC are likely to be too high. As a result of a more realistic conceptual model of uranium waste behaviour, backed up by site observations, and a representation of small scale heterogeneities, the rate of release of uranium from the trenches could be several orders of magnitude lower than previously modelled. This result, combined with the lower

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overall uranium inventory within the trenches, is likely to reduce the risk associated with uranium for the groundwater pathway.

The potential for colloids and organic complexation to increase the mobility of radionuclides within the near field has been assessed as low. Near field colloids do not appear to be stable within the geosphere environment and so colloidal transport of radionuclides from the near field is unlikely to be significant. Modelling studies also indicate that the complexation of natural organic material with uranium, for example, is low and therefore unlikely to have a significant effect on near field solubility or sorption.

The work also confirms that the alkaline conditions associated with the vault waste form will persist for several thousand years. These alkaline conditions are buffered by the dissolution of cement phases within the backfill grout and high pH conditions are predicted to be maintained, despite the accelerated dissolution of these phases due to the degradation of organic material within the waste.

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6 Engineering Performance Assessment

6.1 Introduction

In order to support the development of the radiological safety assessment (RSA) presented as part of this submission, it is necessary to provide an understanding of the nature and evolution of the engineering system that will be present at the LLWR following the withdrawal of control over the facility. The engineering system is referred to as the Single Option (see Section 2.3), with the derivation of this design option described in more detail in Carpenter and Proctor (2007). The work presented in this section is focused on understanding the performance of this currently defined engineering system. Further discussion of the strategic options relating to the post closure engineering design is presented in Volume 2.

Therefore, as part of the LLWR Lifetime Project, an Engineering Performance Assessment (EPA) has been undertaken in order to demonstrate that:

- The engineered system and its impact on the disposal system are understood and represented appropriately in assessment models;
- The natural evolution of the engineered system is understood and represented appropriately in assessment models;
- The role of the engineered system in preventing intrusion scenarios is understood and represented appropriately in assessment models;
- Key engineering uncertainties are addressed; and
- Parameters used for engineering material properties and near field flows are robust and justifiable.

The following sections summarise the progress that has been made in fulfilling these requirements. The EPA has involved a range of tasks aimed at developing a conceptual model of the engineering system, an assessment of the performance of selected components of the system and an evaluation of the groundwater flows within the near field environment.

6.2 Summary of the near field engineering system

The underpinning basis of the current EPA is the specification and design of the future engineering envisaged for the site. As described in Section 2, a design option termed the Single Option has been developed (Carpenter and Proctor, 2007; Belton, 2007). The Single Option describes current and future site management (including both the vaults and the trenches) and closure features (e.g. cap and cut-off wall) that are consistent with the proposed design concept. The main engineering features at the LLWR following withdrawal of control over the facility require representation in the EPA. These features include the trenches, Vault 8 , Vault 9 and the proposed future vaults, final cap, cut-off wall; and vertical drain.

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6.3 Conceptualisation of the near field engineering system

6.3.1 Overview

A conceptual model of the existing and future near field engineering is shown in Figure 6.1, with six main components:

- cap;
- trenches;
- vaults;
- cut-off wall;
- vertical drain; and
- associated geology (i.e. the remnants of the natural geology of the site).

The main interfaces for groundwater flow between the near field and the geosphere are identified in Figure 6.1 as ‘gates’ 1 to 7, representing, for example infiltration through the cap or flows through the vertical drain. It is worth noting that Gate 6 (uncontrolled overtopping or bathtubbing) is included in Figure 6.1 for completeness (i.e. as a theoretical pathway for flow) only and is not considered in the EPA. The vertical drain is included in the current reference design to minimise the likelihood of bathtubbing and it has been assumed that the vertical drains would function well and as designed (see Section 6.3.8).

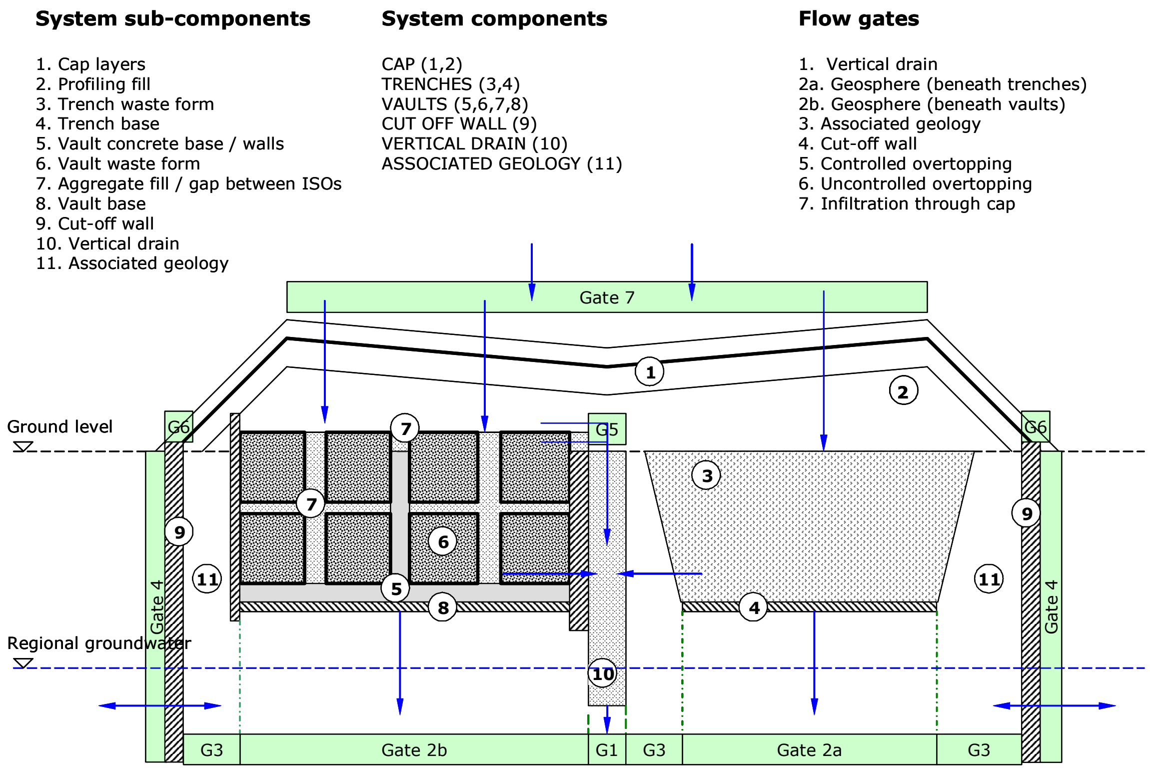

Figure 6.1 Conceptual model of near field engineering and identification of main flow gates

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In implementing this conceptual model, the following processes and uncertainties have been identified.

- Infiltration – a combination of precipitation and cap performance, both subject to change over time due to climate change;
- Location and volumetric rate of lateral flows into and out of the near field;
- Vertical flow (leakage) through the base liner; and
- Component degradation - the performance of the engineered materials will change over time in response to physical, chemical and biological processes.

Component degradation and overall cap performance were evaluated through expert elicitation within an EPA workshop (Paksy, 2008), to provide parameter values to assess impacts on lateral and vertical flows using a near field water balance model. The components included within the elicitation exercise are listed in Table 6.1.

Table 6.1 List of components considered at workshop for elicitation.

Component	Elicited ?	Comment
Cap	Yes	The workshop discussed interim results from a cap assessment study including proposed values of hydrologically effective rainfall. Percolation through the cap liner was elicited at the workshop.
Trench Base	Yes	
Vaults Base	Yes	Assessed separately for Vault 8 and future vaults. Future vaults and Vault 9 are considered to be the same in this respect.
Cut-Off Wall	Yes	
Vertical Drain	No	Anticipated performance discussed briefly in qualitative terms, but performance not elicited due to assumption of sufficient capacity at all times.
Associated Geology	Yes	
Trench Waste Form	No	Not elicited due to assigned low priority.
Vault Waste Form	No	Not elicited due to assigned low priority.
Vault Walls	No	Not elicited due to assigned low priority.

Full details of the elicitation exercise are provided in Section 4 of Paksy (2008), along with the numerical parameters determined. These parameters were provided as to represent upper /

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lower bound and best estimate performance. Components that were not elicited during the workshop were assigned values subsequently, mainly based on values used within the 2002 PCSC (Paksy, 2008).

Key features of the elicitation process are summarised in the sections below.

6.3.2 Cap liner

An assessment of the cap performance was undertaken prior to the elicitation workshop (Thorne, 2008), with this work providing the basis for subsequent elicitation. The expected hydrologically effective rainfall (HER) for the cap is 150 to 400 mm y^{-1} (Thorne, 2008), which is significantly less than that assumed in the 2002 PCSC (560 mm y^{-1}, BNFL, 2002f). This new HER was derived through taking account of both seasonal (monthly) and climatic variations, although explicit consideration of the variability of HER and subsequent sensitivity analysis has not been undertaken during the current work. The likelihood of increased permeability through the clay due to cracking was deemed low. Initial performance (0 – 50 years) is presumed to be controlled by the HDPE geomembrane. Settlement was considered the main mechanism controlling the overall degradation of cap performance over a timescale of a few hundred years, with three timesteps suggested: 0 to 50 years, 50 to 300 years and 300 to 1000 years, with settlement mostly being completed by 300 years. However, it was considered that even after settlement, the performance of the cap would not be affected to a degree that is greater than the uncertainty assigned for the pre-settlement performance.

6.3.3 Trench base

The vertical conductivity of the trench base is assumed to be consistent with that used for recent site scale flow modelling (see Volume 4), with a value of 2E-9 m s^{-1} adopted as best estimate, with the assumption that this value will remain constant over the period of the assessment.

6.3.4 Vault 8 base

The Vault 8 base includes a single layer of bentonite, added to fill in the natural geology where clay was absent. This is therefore similar to the situation within the trenches and so the same hydraulic conductivity as for the trenches was chosen. The same assumption was also made that the hydraulic conductivity will not change over time.

6.3.5 Future vaults base

Unlike Vault 8, the base of the future vaults consists of an upper layer of concrete, a layer of geotextile and then two layers of bentonite (each 0.5 m thick) separated by a layer of granular material. The performance was assumed to be bound by the design criteria of the bentonite after swelling (1E-10 m s^{-1}) and the hydraulic conductivity of the underlying geology, although it was assumed that the vault base would always remain less conductive than the geology. A best estimate (5E-10 m s^{-1}) was chosen to be the mean of the upper and lower bound values.

6.3.6 Cut-off wall

Although the technique for construction is still to be decided, the proposed cut-off wall design (Carpenter and Proctor, 2007) specifies the use of bentonite slurry and/or cement. The

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performance was therefore based on the properties of cement bentonite cut-off walls and bentonite slurry walls obtained from the literature. Degradation of the cut-off wall is presumed to occur over a 500 year period and is represented by a linear increase in the logarithm of the hydraulic conductivity.

6.3.7 Associated geology

The 'associated geology' was assumed to be identical to the natural geology featured in the supporting site-scale groundwater flow model (Volume 4). These values are 2E-9 ms^{-1} for vertical and 1E-5 ms^{-1} for lateral hydraulic conductivity (Paksy, 2008).

6.3.8 Vertical drain

The vertical drain is included in the current reference design to minimise the likelihood of "uncontrolled overtopping (or bathtubbing). As a basis for the current analysis, it was assumed that the vertical drains would function well and as designed. It is possible that the vertical drains could degrade or clog so that this is not the case. For example, as part of the 2002 PCSC it was considered that the performance of the vertical drain (which had a different design to the current vertical drains) would decline after about 100 years, with a reduction of approximately two orders magnitude in hydraulic conductivity (BNFL, 2002f, Table 24). The manner in which the vertical drains, or other engineered features, are designed in order to achieve the desired objectives remains a question for design optimisation (and is discussed in Volume 2).

Thus, for the present work, it was assumed that the vertical drains will have sufficient capacity at all times and so elicitation of this parameter was not required. Performance of the vertical drains was therefore not discussed in detail at the workshop but it was recognised that the values used in the 2002 PCSC could be used for sensitivity analysis as required.

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6.4 Near field flow model

The impact of near field engineering on near field flows was investigated by means of a water balance model, using parameters derived during the elicitation process described above. This model (the near-field water balance model) was based on the conceptual model of the engineered system (Figure 6.1), and calculates volumetric flows through the near field / geosphere gates. This model was implemented using GoldSim version 9.6 (GoldSim Technology Group LLC, 2007). Two model runs were conducted for a reference case and six model runs were conducted to investigate parameter sensitivities.

The near field water balance model was constructed to be consistent with the site-scale groundwater flow model (Arthur *et al.*, 2008). In particular, the groundwater head values were taken directly from this work.

Full details of this modelling are provided in Section 5 of Paksy (2008), which identified that the key features influencing flows through the near field were:

- Percolation through the cap to the waste;

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- Hydraulic conductivity of the trench and vault base;
- Hydraulic conductivity of the cut-off wall and the vault walls (whichever is smaller); and
- Heads in the Upper groundwater outside the cut-off wall.

The best estimate model results suggest that the trenches will remain unsaturated at all times. Given this, and the fact that modelled heads in the Upper groundwater remain below the base of the trenches, there are no horizontal flows into or out of trenches. Therefore, the best estimate model considers that only vertical flow will occur from the trenches through the base. However, sensitivity analyses aimed at investigating parameter uncertainties suggest that the model is very sensitive to the heads in the Upper groundwater and trench base performance. For example, if the head in the trenches increases due to better than elicited “best estimate” performance of the trench base, the leachate flows laterally through the trench walls to the associated geology (and cut-off wall). Such flows are therefore sensitive to the heads in the Upper groundwater and the trench base permeability (and also cap percolation). This situation could arise within the range of parameter uncertainties for the Upper groundwater heads and trench base performance, which would lead to the possibility of additional horizontal pathways from the trenches to the south-east and north-east.

In contrast, the future vaults would maintain a head of about 0.5 to 1m above the liner throughout the assessment period, which would drive a comparatively small horizontal flow through the vault wall to the vertical drain, and a small horizontal flow to the south-east through the vault wall and the cut-off wall.

The picture is more complex for Vault 8, for which the vault walls were assumed not to outperform the cut-off wall. Poor performance of the Vault 8 side walls (and of the cut-off wall after about 500 years) together with high heads in the Upper groundwater on the south-western side of Vault 8, results in high rates of inflows to Vault 8 from the south-west. Groundwater heads to the south west of the disposal area are elevated due to the surface topography in that area. Inflows through the cut-off wall to Vault 8 mainly discharge to the vertical drain through the Vault 8 wall to the north-east and through the associated geology with a lesser discharge to the north-west as horizontal flow. Given the conceptual model assumptions, this could lead to very significant flows towards the vertical drain from Vault 8. Such recharges could impact on the Regional groundwater by leading to development of localised groundwater high below the vertical drain. The possible size of a mound would depend on the relative rates of discharges from the vertical drain compared with normal recharge.

Comparing the relative sizes of different flows through the trenches, vertical flows are more significant than horizontal ones. The opposite is true for Vault 8, given the scenario of high Upper groundwater heads around Vault 8. For Vault 9 and the future vaults, horizontal flows to the vertical drain are comparable to vertical flows through the base. Horizontal flow from the future vaults to the south-east is more than an order of magnitude smaller than flow through the base. Therefore, for the reference case, it may be concluded that vertical flows out of the

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near-field dominate, with the only horizontal flow potentially needing consideration being the flow from Vault 8 to the north-west.

6.5 Summary

As part of the current LLWR Lifetime Project, a programme of work has been undertaken to provide data on the anticipated performance of the engineered barrier system and its impact on leachate movement within the near field and releases from the near field to the geosphere.

The work presented in this volume has focussed on the new design option for the LLWR engineered barrier system, which has some differences to the post-closure engineering design assumed within the 2002 PCSC. Further discussion of the strategic options relating to the post closure engineering design is presented in Volume 2.

The main output from this work is the derivation of a near field water balance model, based on a conceptual model of the engineered system, which has calculated volumetric flows through the near field system. Key findings from this work include:

- The trenches will remain unsaturated throughout the assessment period, with the only flow from the trenches being vertical flow through the base.
- Within the future vaults environment, some horizontal flow through the vault walls to the vertical drain is estimated, due to the maintenance of a head of about 0.5 to 1m leachate above the liner. These horizontal flows to the vertical drain are generally comparable in size to vertical flows through the base.
- Significant horizontal flows into the vertical drain from Vault 8 through the side wall are estimated. This is due to the relatively poor performance of the Vault 8 side walls, together with high heads in the Upper groundwater on the south-western side of Vault 8, resulting in high rates of groundwater inflow to Vault 8 from the south-west.

Overall, it may be concluded that vertical flows out of the near-field dominate, with the only significant horizontal flow being the flow from Vault 8 to the north-west, which will occur only after about 500 years as the engineering degrades. However, the near field water balance model is informed by the site-scale groundwater flow model, and within the range of parameter uncertainties associated with this model allied to uncertainties in trench base performance (within the range considered in the EPA), there is a possibility of significant additional horizontal flows from the trenches.

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7 Representation of the Near Field Within the Assessment Model

7.1 Introduction

The present document is one of a set of 5 volumes that is designed to satisfy Requirement 2 of Schedule 9 and provide an assessment of the radiological impacts from the trenches and vaults and an assessment of radiological capacity. A number of assessment calculations are included within this submission, encompassing groundwater, gas and human intrusion pathways.

In developing these assessment calculations, it is necessary to define a range of conceptual models describing how the disposal system is expected to evolve and to describe how the key components of the disposal system will be represented in the safety assessment.

As part of the 2002 PCSC, a detailed analysis of the key features, events and processes (FEPs) was undertaken, including the derivation of an LLWR-specific FEP list (using an interaction matrix approach). A simplified analysis of FEPs, compared with the relatively complex treatment adopted in the 2002 PCSC, is proposed for the future assessments. This acknowledges that significant understanding has been developed through the 2002 OESC and PCSC analyses. The approach for the current radiological safety assessment is to develop conceptual models through focus on the key FEPs identified from the 2002 OESC and PCSC, and from work that has been undertaken since 2002, including that within the LLWR Lifetime Project.

This section presents an overview of the development of a conceptual model for the near field, based on the discussions of the individual components of the system described in the previous sections. The focus of this section is the near field in relation to the groundwater pathway, which requires a reasonable level of complexity.

In contrast, other pathways considered within this submission (including future human actions, gas pathways and disruption of the site through coastal processes) only require a simpler conceptual model of the source term. For example, a simplified approach has been adopted in which the source term concentrations are derived to account for radioactive decay and ingrowth, but losses due to leaching and transport of radionuclides are not included. This is a conservative approach with respect to calculation of impacts associated with these pathways, which are related to the residual activity remaining in the near-field. Further details of these pathways can be found in Volume 5 and the underpinning references (e.g. Galais, 2008 and Ball *et al.*, 2008).

The mathematical representation of the conceptual models in the assessment codes is not included within this report, but is fully documented in Volume 5 of this submission.

7.2 Assessment time period

The timescales of interest for the assessments presented within this submission is of the order of a few thousand years. This is the timescale over which gross disruption of the wastes by

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coastal erosion is considered to occur (see Volume 4). Therefore, the majority of assessment calculations will be undertaken over the time period up to around 5,000 years After Present (AP). Where impacts are shown to demonstrate an increasing trend at 5,000 years AP, calculations may be undertaken for an extended time period.

7.3 Near field conceptual model for the groundwater pathway

7.3.1 Introduction

For the analysis of impacts by release of radioactivity to groundwater, the near field essentially represents the source component of the source-pathway-receptor linkage considered in safety assessments. This section provides an overview of the conceptual model for the near field in relation to the groundwater pathway. A key requirement is the need to represent and define the geometries and dimensions of the near field features being represented flow, transport and chemical characteristics of the wastes, engineering and pathways in the near field and how these change with time.

In order to define the FEPs to be included within the near field conceptual model, the 2002 PCSC (BNFL, 2002b) has been used, supplemented by the subsequent work undertaken within the LLWR Lifetime Project (and summarised in the Sections above).

The key FEPs included in the conceptual model for the near field are listed below.

Trench disposal facility:

- The inventory of radioactive wastes disposed to the trenches and radioactive decay of the inventory.

- Inputs and outputs of water through the trench wastes, leading to leaching of radionuclides. Inputs are from infiltration of water through the cap and inflow of groundwater laterally through the cut-off walls (initially expected to be minimal inflow with an increase with time as the engineering degrades). Outputs are controlled by the performance of the closure engineering.

- Solubility limitation for some radionuclides, with variation in time and space due to transient biogeochemical processes.

- Retardation of radionuclide transport via sorption processes onto waste materials (e.g. soils present in the trenches) and engineering materials (e.g. bentonite).

- Immobilisation of uranium within a resistant matrix (waste form), where the release of uranium is limited by the dissolution behaviour of the waste form (fluoride residues).

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Vault disposal facilities:

- Inventory of radioactive wastes disposed to Vault 8 and for potential disposal to Vault 9 and future vaults, and radioactive decay of the inventory.

- Inputs and outputs of water through the vault wastes, leading to leaching of radionuclides. Inputs are from infiltration of water through the cap and inflow of groundwater laterally through the cut-off walls and vault walls (initially expected to be minimal inflow with an increase with time as the engineering degrades). Outputs are controlled by the performance of the vaults and closure engineering.

- Solubility limitation for some radionuclides under alkaline conditions established in the cementitious vault waste.

- Retardation of radionuclide transport via sorption processes onto waste form materials (e.g. cementitious materials).

7.3.2 Inventory

An updated inventory has been provided as part of the LLWR Lifetime Project (Wareing *et al.* 2008) and is summarised in Section 4. As a result of this update and subsequent screening of the inventory, the following radionuclides are currently included within the near field model:

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, Mo-93, Nb-94, Nb-95, Ni-63, Np-237, Pa-231, Pb-210, 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.

Screening of radionuclides was based on that performed in the 2002 PCSC with, in addition, a review of the new inventory and identifying any radionuclide with a significantly increased activity within the 2007 inventory; screening out radionuclides that were insignificant for the groundwater pathway within a 2500 years assessment time from the 2002 PCSC; and identification of radionuclides with high seafood concentration factors in the marine environment.

The inventory contains detailed information concerning the locations of particular radionuclides within the disposal system; information that has been used extensively in building up the detailed underpinning geochemical models presented in Section 5. However, within the current assessment conceptual model a simplified, more homogeneous representation of the wastes is considered appropriate. Separate source terms are used to represent different parts of the disposal facility, based on distinctions in the inventory, biogeochemistry and flow paths.

The discussions presented earlier indicate that the nature of specific uranium disposals is an important feature to represent within the trench environment. Therefore four main regions are envisaged:

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- Regions of the trenches associated with fluoride-type solubility control for uranium – mainly at the northern end of trenches 1, 4, 5 and 6 (Section 6.3 of Small *et al*., 2008a);

- Trench region 6.1 with fluoride-type solubility control for uranium associated with higher concentrations of fluoride and uranium in leachate (Section 6.1 of Small *et al*., 2008a);

- Regions of the trenches associated with the disposal of other residues from the Springfields site, including acid leached yellowcake and scrap material; and

- Other regions of the trenches with a U(IV) or U(IV) solubility concept as previously considered in the 2002 PCSC.

Further discussions of these regions in relation to the representation of uranium behaviour are presented later.

The vault environment also consists of three regions but these are related to the specific disposal facilities in order that the required endpoints can be determined according to these regions, to inform management decisions regarding existing facilities and future facilities, thus:

- Vault 8;

- Vault 9; and

- Future vaults

The inventory of each region is assumed to be uniformly distributed throughout the region and the spatial variation of flows within each region assumed to be negligible. A schematic representation of near field is provided in Figure 7.1

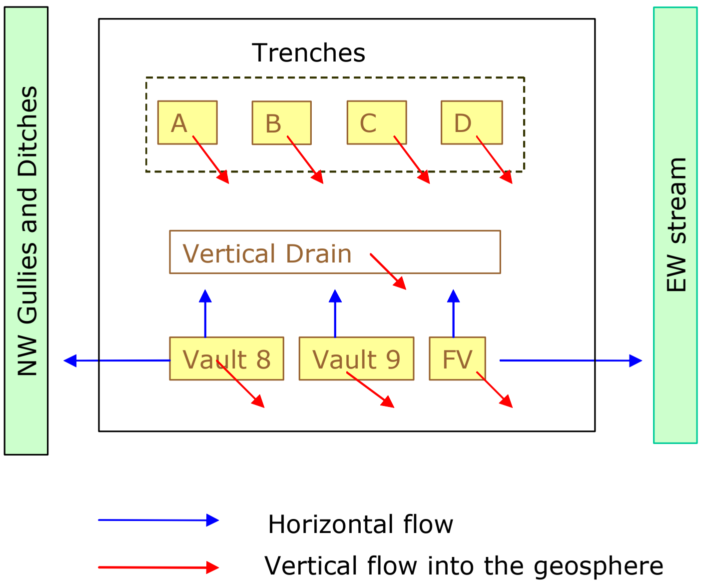

Figure 7.1 Schematic representation of the near field

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7.3.3 Near field engineering and water flows

The representation of near field flows is informed by the results of the Engineering Performance Assessment presented in Section 6. Flows into and out of the near field are assumed to be either in a vertical or lateral (horizontal) direction. Flows out of the near field model are represented in Figure 7.1. Although vertical flows are assumed to dominate, there are significant horizontal flows from Vault 8 and smaller horizontal flows from the future vaults that require representation.

7.3.4 Biogeochemical evolution

The 2002 PCSC (BNFL, 2002c) showed that the release of many radionuclides is dependent on the prevailing biogeochemical conditions, for example uranium demonstrates a marked difference in solubility for reduced and oxidised conditions in the trenches. It is therefore a requirement that this potential variation is represented within the near field model.

The 2002 PCSC (BNFL, 2002c) indicates the following broad characteristics of the trenches and vaults.

- Trench waste reducing conditions: 0 – 4000 years;
- Trench waste oxidising conditions: 4000 + years; and
- Vault waste reducing conditions, high pH: throughout

Recent work on the near field biogeochemistry (Small *et al*, 2008a; Kwong *et al*, 2008; Kwong, 2008) have indicated that these characteristics remain valid for the current radiological safety assessment. Therefore, it is not considered necessary to explicitly model the development and evolution of biogeochemical conditions within the current model of the near field. Instead, the conditions listed above are assumed to prevail, with parameters describing radionuclide behaviour selected to be appropriate to these conditions.

With reference to the discussion on assessment time periods, as it is anticipated that the site will be disrupted by coastal processes within some thousands of years, there may not be time for the re-oxidation of the trenches to occur.

7.3.5 Radionuclide behaviour

The results of the near field flow modelling indicate the possibility that parts of the near field environment are likely to be at least partially unsaturated. This conclusion obviously has implications to the flow of water into and out of the near field, and the representation of this is shown in Figure 7.1. Another potential implication of the partial saturation of the near field relates to the leaching of radionuclides from waste into groundwater. An unsaturated environment would result in limiting this leaching and so potentially reduce the release of radionuclides into groundwater. However, a cautious approach has been adopted whereby it is assumed that groundwater is in equilibrium with the radionuclide content of the waste and release is not limited by the degree of saturation. The release of radionuclides from the near

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field is therefore assumed to be largely dependent on retardation of radionuclide transport via sorption processes and solubility limitation for some radionuclides.

Retardation processes act to delay the time taken for dissolved species to travel through particular media. These processes include sorption, which can be defined as the accumulation of matter at the interface between the solid surface and the aqueous phase. Sorption includes ion-exchange, where the interaction is controlled primarily by electrostatic attraction, and surface complexation onto variable charge sites.

Within the current near field model it is assumed that sorption processes can be treated as instantaneous and reversible and therefore described by an element-dependent distribution coefficient, K_d, defined as the ratio of sorbed concentration over the concentration in the aqueous phase. Therefore, in order to represent sorption processes within the near field, a distribution coefficient appropriate to the prevailing conditions is required for each radionuclide.

In the LLWR trenches, waste was disposed of by loose tipping of waste. There are a variety of solid phases that could potentially act as substrates for sorption in the trenches. Previously (Randall and Keith, 2002) identified the following sorption substrates:

- Soil present as disposed contaminated waste, and present as backfill and fire breaks;
- Metallic corrosion products comprising oxides, hydroxides and carbonates formed by corrosion of mainly iron waste;
- Plastic and cellulose based wastes and their solid degradation products including living and dead biomass and resulting solid humus; and
- Disposed inorganic and mineral wastes including building rubble, concrete and sand. Primary mineral surfaces are here likely to consist of silicates and carbonates of variable grain size.

The latter category could now be considered to include the uranium waste form disposed in the trenches.

In the 2002 assessment the conservative assumption was made that the dominant sorption substrate in the trenches is the disposed soil inventory. The first reason for this choice is that the amount of disposed soil is known (within the range of uncertainty associated with inventory records and calculations) in contrast to the amount of soil backfill for example. Secondly, the mechanisms describing sorption onto soils and sediments are more readily understood and hence more robustly justifiable than for other potential substrates, such as the cellulosic waste or microbial biomass. In order to claim benefit for sorption onto other potential substrates, it would be necessary to quantify the behaviour under LLWR conditions (Small *et al.*, 2008b).

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It is therefore assumed that the dominant sorbant in the trenches is the disposed soil inventory and the fire breaks, therefore sorption in the trenches is conceptualised using K_d values appropriate to soils.

Within the vaults, the cementitious grout used to fill void space between compacted waste pucks has generally good sorption properties for a wide range of radionuclides. Additionally, there are large quantities of soil within the vaults inventory (Section 4) It is therefore assumed that sorption occurs onto the cementitious grout and the disposed soil, with sorption represented using K_d values appropriate to a cementitious environment.

Solubility controls are likely to be only relevant to a few radionuclides. Table 7.1 shows the radionuclides that demonstrated some solubility control within the 2002 PCSC.

Table 7.1 Summary of solubility controlled radionuclides from the 2002 PCSC (Section 6.5.1 of BNFL, 2002c)

Phase	Duration (y)	Location	Controlling influences
$Np(OH)_4$	0-240	Trenches	pH dependent solubility, in some trench cells initial concentration (approximately 1E-9 mol l^{-1}) is close to solubility limit. Solubility control is short lived and is lost because of advective flow.
$Th(OH)_4$	Throughout	Throughout	Constant solubility under most chemical conditions but increased by acetate complexation during initial 1,000 years.
UO_2	0-3,500	Trenches	Solubility controlled by reduced conditions associated with waste degradation and corrosion. Uranium dissolves as site oxidises back to groundwater conditions.
$CaUO_4$	Throughout	Vaults	This uranium (VI) phase maintains solubility control under alkaline conditions in the vaults. In some vault cells solubility control is eventually lost (> 5,000 years) when cement phases react out increasing uranium solubility through carbonate complexation and by decreased pH.
PbS	Throughout	Throughout	In the presence of sulphide, lead is solubility controlled. The DRINK model only considers active Pb-210 which is maintained as a decay product from uranium and thorium.
TcO_4	0-450	Throughout	Technetium is initially solubility controlled when strongly reduced conditions (fermentation couple) are established in the trenches. In the vaults technetium is solubility controlled in Vault 8 and future Vaults 9 to 22 as a consequence of the initial technetium inventory and the initial pH and pe conditions.

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Based on the 2002 PCSC, only uranium and, thorium are presumed to be solubility controlled within the current conceptual model of the near field. As discussed in Section 3 and Section 6, changes to our understanding of the climate and landscape evolution of the site and the degree of saturation within the near field may have some influence on the evolution of the site and the behaviour of radionuclides. Therefore, at this stage of the Lifetime Project it is assumed that solubility limits are to be applied for thorium according to the findings of the 2002 PCSC summarised in Table 7.1. Neptunium, lead and technetium have not been included at this stage either because the duration of solubility control is short lived and controlled by advective flow or the solubility control is determined by geochemical conditions (pe, pH etc) that are subject to uncertainty.

The representation of uranium solubilities in the trenches requires some revision as a result of the work undertaken as part of the LLWR Lifetime Project (summarised in Section 5). This demonstrated that the release of uranium will be largely controlled by the dissolution of uranium process residues. It is therefore appropriate to represent the solubility of uranium according to the following categories.

- Uranium solubility in regions of primary fluoride type solubility control. Uranium process residues, including magnesium and calcium fluoride residues, comprise an important component of the trench uranium inventory. The concept for this fluoride residues waste form is that uranium is released in proportion to the mass of fluoride residue that dissolves in groundwater (Small *et al*, 2008a). Given the mole fraction of uranium within the magnesium and calcium fluoride residues (less than 1% - Section 5), uranium release through dissolution of fluoride is conceptualised as a uranium solubility limit equivalent to the fluoride solubility multiplied by the mole fraction of uranium present within the fluoride waste form.

- Uranium solubility in Trench region 6.1. This region is characterised by higher concentrations of fluoride and uranium in leachate (Sections 6.1 and 6.3 of Small *et al*., 2008a). It is assumed that the uranium solubility in this region is an order of magnitude higher than in the regions of primary fluoride-type solubility control, since the concentrations of fluoride and uranium in leachate are about an order of magnitude higher in this trench region compared with the other regions of fluoride-type solubility control (Sections 6.1 and 6.3 of Small *et al*., 2008a).

- Uranium solubility in other areas of the trenches, including Trench 7. Uranium disposals in other areas of the trenches, in particular Trench 7, are not characterised as having the same fluoride association as disposals in Trenches 1, 4, 5 and 6. Under reduced conditions, the solubility of uranium in this region is controlled by the uranium (IV) phase (Section 6.5.1 of BNFL, 2002c).

- Regions of the trenches associated with the disposal of other residues from the Springfields site, including insoluble mineral residues from yellow cake dissolution in hot nitric acid and scrap materials composed of siliceous ceramics and graphite. The mineralogical nature of these materials is unknown, however given the process treatment they are likely to be characterised by low solubility. Therefore, release of

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uranium from these waste forms is assumed to be described by the same solubility limits as for the fluoride residues.

Colloids and organic materials (e.g. complexing agents) are assumed not to have a significant impact on radionuclide transport and are therefore not considered (see discussion in Section 5).

Microbial activity is considered to only impact on radionuclide migration for the groundwater pathway through changes to the ambient geochemistry, e.g. microbial processes have no direct impact on sorption properties.

C-14 represents an exception to the usual sorption and solubility behaviour exhibited by other radionuclides. Stable carbon forms an important part of the biogeochemical reactions occurring within the near field system and so C-14 will be incorporated within these reactions. Within the 2002 PCSC DRINK near field model, C-14 was considered in relation to microbiological and chemical reactions, assuming isotopic fractionation to be insignificant (Section 6.5.1 of BNFL, 2002c). For the current radiological assessment calculations, “pseudo-solubilities” have been derived based on the aqueous concentrations calculated within the 2002 PCSC DRINK near field model.

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8 Summary

The present document is Volume 3 of a set of 5 volumes produced as part of the LLWR Lifetime Project that is designed to satisfy Requirement 2 of Schedule 9 of the new single authorisation granted by the EA (EA, 2006). The focus of this Volume has been to demonstrate the advancements in understanding of key aspects of the near field gained since the submission of the 2002 PCSC.

The document has provided a summary of a number of work streams, all contributing to the near field area. These are:

- Re-evaluation of the disposed inventory;
- Biogeochemical modelling of the near field; and
- Engineering Performance Assessment.

These areas have been combined within an overall conceptual model of the near field.

The derivation of an updated inventory for the LLWR site represents a significant body of work. A significantly greater reliance has been placed on the use of real disposal records rather than the application of backfitting techniques to relate the historical disposals to current waste streams. This has resulted in a good knowledge of the inventories of key radionuclides and, importantly, a detailed understanding of the distribution of this inventory across the disposal system.

These improvements have been evident in the re-evaluation of the potential impacts of disposed uranium within the trenches, one of the key radionuclides within the 2002 PCSC. The re-evaluation of the inventory of disposals at the LLWR, utilising historical disposal records, has resulted in a reduction in the trench uranium by a factor of two, with the vast majority (over 95%) of this uranium inventory associated with disposals from the Springfields site. An increased understanding of the nature of this waste and subsequent geochemical modelling of uranium release from the trench environment has shown that the uranium fluxes from the trenches calculated as part of the 2002 PCSC are conservative. As a result of a more realistic conceptual model of uranium waste behaviour, backed up by site observations, and a representation of small scale heterogeneities, the rate of release of the majority of uranium from the trenches could be several orders of magnitude lower than previously modelled.

The inventory re-evaluation has also resulted in a reduction in the overall trench radionuclide inventory by around 20% from that calculated for the 2002 Safety Cases and a much more detailed representation of the heterogeneity of the disposals, to the level of sub-bays. This information is important for subsequent modelling and the management of potential optioneering solutions.

For the future vaults it has been shown that between 70% and 90% of activity is represented by just five key waste streams, several of which are common to more than one group. In

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particular, just one Springfields decommissioning waste stream with a volume of approximately 3,000 m^3 contributes almost 90% of future uranium activity and almost all future Np-237 activity.

The work presented within this Volume also confirms that the beneficial alkaline conditions associated with the vault waste form will persist for several thousand years.

A detailed evaluation of the performance of the near field engineering has also been undertaken. The importance of these engineering features is considered to have increased since the 2002 PCSC, given that the assessment timeframes are much shorter before disruption of the wastes by coastal erosion. The engineering performance assessment has identified that, in contrast to the 2002 PCSC, the post-closure engineering features will continue to function over the time period of interest and therefore play a significant role in limiting flows into and out of the facility.

As a result of this engineering performance assessment, a near field water balance model has been produced to represent the flow of groundwater through the near field environment, taking into account the site scale groundwater model (see Volume 4) and the behaviour and properties of the near field post closure engineering.

This model indicates that, in general, vertical flows out of the near-field dominate, with the only significant horizontal flow being the flow from Vault 8 to the north-west. However, within the range of parameter uncertainties associated with modelled groundwater heads and trench base performance, there remains the possibility of significant additional horizontal flows from the trenches.

This information has been combined with the key messages from the 2002 PCSC to produce an updated conceptual model of the near field.

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