

Evaluation of Colloid Transport Issues and Recommendations for SKI Performance Assessments

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This report concerns a study which has been conducted for the Swedish Nuclear Power Inspectorate (SKI). The conclusions and viewpoints presented in the report are those of the authors and do not necessarily coincide with those of the SKI.

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Summary

Background and Objectives

Colloids are minute particles in the size range 1 nm to 1 μ m that can remain suspended in water. Groundwaters often contain significant populations of natural colloids, and these can interact with pollutants and influence their transport. In radioactive waste disposal systems, natural colloids and waste- and repository-derived colloids may influence radionuclide transport. Recent observations made by United States Department of Energy (DOE) researchers at the Nevada Test Site (NTS) have demonstrated migration of Pu 1.3 km from the site of the BENHAM underground nuclear tests, and suggest that the migration may have occurred owing to the attachment of the Pu to colloids.

The primary objective of this project was to develop recommendations to SKI for evaluating the potential significance of colloids in performance assessment (PA) studies by:

- Updating a previous review of the treatment of colloids in PA studies for radioactive waste repositories, to include information on PAs published in the period 1997-1999.
- Reviewing Svensk Kärnbränslehantering's (SKB's) work on colloidal radionuclide transport.
- Reviewing the observations of radionuclide transport at the NTS.

Colloids in Performance Assessment

We have reviewed sixteen PAs conducted in nine countries and have made the following observations:

- All PAs include colloid formation and colloid transport in their FEP list. Although some programmes have deferred consideration of colloidal radionuclide transport until further research has been performed, more recent PAs do account for the effects of colloids.
- PAs of disposal systems in which the waste canister is surrounded by a bentonite buffer do not consider the effect of colloids on the source term. These PAs assume that all colloids are filtered by the bentonite buffer and cannot escape from the near-field. These PAs all assume that quality control measures will ensure adequate emplacement of the buffer, and that the buffer will perform as expected. Failure of the bentonite buffer to filter colloids is not considered as an alternative scenario.
- PAs of disposal systems with no bentonite buffer have to account for mobilisation of radionuclides from the waste by colloids. The concentration of colloids that may form in the repository is a key uncertainty.

- Many PA programmes have modelled colloidal radionuclide transport in the geosphere using one-dimensional transport equations. No PA has included a comprehensive treatment of colloid transport using mechanistic modelling. Some PAs have not undertaken modelling of colloid transport in the geosphere, but have relied instead on arguments that such transport processes will be of low consequence to the performance of the disposal system.
- Five PAs have shown the effects of colloid transport through the geosphere to be potentially significant (NAGRA's Kristallin-I, Japan's H-12, HMIP's Sellafield Assessment, Nirex's Nirex 97, and the US DOE's TSPA-VA for Yucca Mountain).

SKB's Treatment of Colloids

For the disposal of spent nuclear fuel, SKB has developed the KBS-3 concept. In the KBS-3 concept, SKB plans that after 30 to 40 years of interim storage, spent fuel will be placed in copper canisters, surrounded by a bentonite buffer, and disposed at a depth of about 500 m in crystalline bedrock.

For the disposal of long-lived low-level and intermediate-level radioactive waste, SKB has developed the SFL 3-5 concept. In the SFL 3-5 concept, waste is placed in underground disposal caverns and surrounded by cementitious materials and crushed rock.

Colloidal radionuclide transport is unlikely to be significant to the performance of the KBS-3 concept for the disposal of spent fuel. The KBS-3 concept includes a bentonite buffer that is likely to prevent the release of waste- and repository-derived colloids from the repository. Saline groundwaters may provide an additional barrier to colloidal radionuclide transport, if the repository is sited at a location where groundwaters in the host rock are sufficiently saline that they destabilise colloidal species. However, further work is needed to consider the effects of waste- and repository-derived colloids in calculations of the amount of radionuclides that may be transported to the biosphere following an earthquake that damages the engineered barriers, or a future borehole that inadvertently intersects the waste.

The potential significance of colloidal radionuclide transport to the SFL 3-5 concept for the disposal of low-level and intermediate-level waste is uncertain. In contrast to the KBS-3 concept, the SFL 3-5 repository does not incorporate a bentonite buffer and there is a greater chance that waste- and repository-derived colloids may contribute to radionuclide transport. In addition, when compared to the KBS-3 concept, the SFL 3-5 repository will contain a greater range of waste materials from which colloids may be derived. Furthermore, some of these colloid generation processes may occur to a greater extent in the SFL 3-5 repository than in the KBS-3 repository.

Radionuclide Transport at the Nevada Test Site

Three Pu transport mechanisms may have operated, subsequent to the BENHAM test at the NTS:

- Prompt injection of fission product material, either as a plasma, or in gaseous or liquid form, along fractures. However, prompt injection has never been observed to operate over distances greater than 300 m from the site of other NTS nuclear tests.
- Transport of dissolved Pu species in groundwater. However, it is likely that most Pu would be sorbed onto fracture surfaces.
- Colloidal transport of Pu. Co, Cs, Eu and Pu were observed to be associated with colloids in the NTS groundwaters. However, the distance over which the Pu was transported with colloidal material remains unknown.

Many uncertainties remain in understanding of radionuclide migration and colloid transport at the NTS, including:

- The radionuclide transport pathway is unknown.
- The maximum extent of Pu migration is unknown. Insufficient boreholes have been drilled to determine the current spatial distribution of Pu from the BENHAM test.
- It is not known if migration of Pu is unique to the BENHAM test.
- It is not known how the radionuclides are associated with colloids, or whether the Pu exists as intrinsic colloids or pseudocolloids.

As a result of these observations and uncertainties, the Yucca Mountain Project has developed a relatively sophisticated treatment of colloids in their recent PA, which is discussed in detail in this report. The PA is notable because both reversible and irreversible sorption of radionuclides to colloids was included in PA calculations, and dose comparisons made.

Comparison between NTS Observations and SKB's Waste Disposal Concepts

At the NTS, colloids may have promoted ≥ 1.3 km migration of Pu and other radionuclides in 30 years. There are significant differences between SKB's waste disposal concepts and the NTS environment. The key difference is the contrasting mode of emplacement of the radioactive material. In SKB's waste disposal concepts, the waste is emplaced passively and is protected from the surrounding environment by an engineered barrier system. At the NTS, the radioactive source material was emplaced explosively, with attendant fracturing of the surrounding rock, and was immediately exposed to groundwater. In SKB's waste disposal concepts, the waste could only ever become directly exposed to groundwater after the engineered barriers were breached.

Recommendations

The project findings lead us to make the following recommendations:

- Further work should be conducted to assess the potential significance of colloids to the performance of SKB's concepts for radioactive waste disposal. At the same

time, SKB's work on colloids should be kept under review. The work should be directed at assessing the potential significance of colloids to the performance of both the KBS-3 disposal concept, and the SFL 3-5 disposal concept, in which there remain large uncertainties related to colloids.

- Benefit can be gained by monitoring progress in international collaborative work on colloids.
- A "watching brief" should be maintained on future developments at the NTS. New results on NTS radionuclides and colloids will be available shortly from DOE's laboratory studies, field experiments and modelling programmes.
- Important lessons may be learned by continuing to monitor developments in other PA programmes, and in international collaboration work on colloids.

Confidence can be built in regulatory assessment programmes by the conduct of independent calculations; regulators should consider the conduct of colloid transport calculations and the necessity of incorporating colloidal effects in PA.

Evaluation of Colloid Transport Issues and Recommendations for SKI Performance Assessments

1 Introduction

Svensk Kärnbränslehantering AB (SKB) has responsibility for disposal of Sweden's radioactive waste. For the disposal of spent nuclear fuel, SKB has developed the KBS-3 concept. In the KBS-3 concept, SKB plans that after 30 to 40 years of interim storage, spent fuel will be placed in copper canisters, surrounded by a bentonite buffer, and disposed at a depth of about 500 m in crystalline bedrock. For the disposal of long-lived low-level and intermediate-level radioactive waste, SKB has developed the SFL 3-5 concept. In the SFL 3-5 concept, waste is placed in underground disposal caverns and surrounded by cementitious materials and crushed rock.

Colloids are minute particles in the size range 1 nm to 1 μ m that can remain suspended in water. Groundwaters often contain significant populations of natural colloids, and these can interact with pollutants and influence their transport. In radioactive waste disposal systems, natural colloids and waste- and repository-derived colloids may influence radionuclide transport.

Early attempts to include colloids within performance assessments (PAs) for radioactive waste disposal systems were limited. This was a result of:

- Limited understanding of the numerous physico-chemical processes involved in colloid formation and transport, and of the couplings between these processes.
- A lack of relevant data to describe colloidal processes.
- Limited capability to simulate colloidal processes using appropriate models.
- The need for PA models to be relatively simple and computationally efficient.

Since the early 1990s, however, there has been a growing awareness of the potential importance of colloids, and as greater computing resources and more data have become available, PA models have increasingly included colloids in quantitative analyses of repository safety.

Recent observations made by United States Department of Energy (DOE) researchers at the Nevada Test Site (NTS) have demonstrated migration of Pu 1.3 km from the site of the BENHAM underground nuclear test; the researchers suggest that the migration may have occurred owing to the attachment of the Pu to colloids (Kersting *et al.*, 1999).

The primary objective of this project was to develop recommendations to SKI for evaluating the potential significance of colloids in PA studies by:

- (i) Updating a previous review of the treatment of colloids in PA studies for radioactive waste repositories (Bennett *et al.*, 1999) to include information on PAs published in the period 1997-1999.
- (ii) Reviewing SKB's work on colloidal radionuclide transport.
- (iii) Reviewing the observations of radionuclide transport at the NTS.

The report is organised as follows:

- Sections 2, 3 and Appendix A address Objective (i). Section 2 provides a summary of the treatment of colloids in recently published PAs. Section 3 summarises the treatment of colloids in SKB's PAs, and in the recent Yucca Mountain Viability Assessment (TSPA-VA). We focus on the TSPA-VA here because in this PA, special attention was paid to the treatment of colloids given the observations from the NTS. Appendix A presents detailed reviews of the treatment of colloids in recent PAs in Finland, Japan and the UK.
- Sections 4 and 5 address objective (ii). Section 4 provides a summary of recent observations at the Nevada Test Site (NTS). Section 5 compares the relevance of the NTS observations with the expected behaviour of colloids in the Swedish KBS-3 disposal concept.
- Section 6 addresses objective (iii) by providing an evaluation of SKB's treatment of colloids in its recent PAs.
- Section 7 addresses objective (iv) and contains our conclusions and recommendations to SKI.

2 Treatment of Colloids in Performance Assessment

2.1 Coverage of Review

In order to provide an overview of the requirements, difficulties, and practicalities of accounting for colloid formation and transport in PA, we have reviewed a representative selection of recently published PA work from the following nine countries with radioactive waste management programmes:

Belgium: Disposal of high-level waste and spent fuel in clay at the Mol site. Second phase of PA published in 1996-1997 (Marivoet *et al.*, 1996, 1997, on behalf of ONDRAF/NIRAS).

Canada: Demonstration of the concept for disposal of spent nuclear fuel in crystalline basement. PA published in 1994 (Concept Assessment; AECL, 1994).

Finland: Evaluation of three sites for disposal of spent nuclear fuel in crystalline basement. Second and third phases of generic PA (TILA-96; TILA-99; Vieno and Nordman, 1996, 1999, on behalf of POSIVA).

Japan: Development of a PA methodology for a hypothetical underground repository (H-3; H-12; PNC, 1992; JNC, 2000).

Netherlands: Disposal of high-level waste in a salt diapir. Preliminary PA for a hypothetical site published in 1993 (PROSA; Prij *et al.*, 1993, on behalf of ECN).

Sweden: Disposal of spent nuclear fuel in crystalline basement. PA methodologies for a hypothetical site published by the regulator (SITE-94; SKI, 1996) and the waste management company (SKB 91; SR-95; SR-97; SFL 3-5; SKB 1992, 1995, 1999a, 1999b).

Switzerland: Disposal of high-level waste in crystalline basement is currently one disposal option. Second phase of PA for a hypothetical site published in 1994 (Kristallin-I; NAGRA, 1994).

UK: Disposal of intermediate-level waste in fractured volcanic basement. Aspects of PA requirements were examined by the regulator in 1992-1994 (Sellafield Assessment; Intera Information Technologies, 1996, on behalf of HMIP). A recent PA by the waste management company UK Nirex (NIREX-97; Nirex, 1997) considered transport of radionuclides by colloids.

USA: Disposal of defence transuranic waste in bedded salt. PA submitted to the regulator in 1996 as part of the Compliance Certification Application (CCA) for the Waste Isolation Pilot Plant (WIPP) (US Department of Energy, 1996). Disposal of spent

fuel in unsaturated volcanic ash-flow tuffs. PA submitted as part of the Viability Assessment for the Yucca Mountain site (TSPA-VA; US Department of Energy, 1998).

2.2 Review Conclusions

2.2.1 Review Findings

Table 2.1 and the following text summarise the key points from our review of the treatment of colloids in recent PAs. Section 3 provides further detail on the treatment of colloids in Swedish PAs, and in the recently published Yucca Mountain Viability Assessment PA. Annex A provides details of the treatment of colloids in recent PAs in Finland, Japan and the United Kingdom. Details of the treatment of colloids in the other PAs are given in Bennett *et al.* (1999).

The main findings of our review of the treatment of colloids in recent PAs are summarised below:

- All programmes include colloid formation and colloid transport in their FEP list.
- PAs of repositories for spent nuclear fuel and HLW that contain a bentonite buffer do not consider the effect of colloids on the source term. These PAs assume that all colloids are filtered by the bentonite buffer and cannot escape from the near-field (e.g., AECL's Concept Assessment, SKB's SR-95 and SR-97, NAGRA's Kristallin-1). There is considerable uncertainty over the scale of colloid generation from waste, glass and bentonite degradation. However, the PAs all assume that quality control measures will ensure adequate emplacement of the buffer, and that the buffer will perform as expected. Failure of the bentonite buffer to filter colloids is not considered as an alternative scenario.
- PAs of waste repositories with no bentonite/clay buffer have to account for mobilisation of radionuclides from the waste by colloids (e.g. HMIP's Sellafield Assessment, US DOE's WIPP CCA and Yucca Mountain TSPA-VA). Experimental observations have been used to document higher effective solubilities associated with radionuclide binding to mineral fragments, microbes, and organic matter. The concentration of colloids that may form in the repository is a key uncertainty.
- Some programmes have deferred consideration of naturally-occurring colloid transport in PA until further research has been performed (e.g., SKI's SITE-94, POSIVA's TILA-96 and TILA-99, PROSA).
- Some programmes have not modelled colloid transport in the geosphere, and have relied instead on arguments that such transport processes will be of low consequence to the performance of the disposal system. Estimates of consequence were made on the basis of theoretical considerations (AECL's Concept Assessment) or on the basis of experimental observations (US DOE's WIPP CCA).

- No PA has included a comprehensive treatment of colloid transport using mechanistic modelling. However, the modelling approach used in NAGRA's Kristallin-I, in the US DOE's TSPA-VA, and the approach for the Culebra at the WIPP site (US DOE's WIPP CCA) could be used to simulate different colloid velocities and colloid host-rock interactions. Also, the colloid transport calculations made in support of HMIP's Sellafield Assessment, Nirex's Nirex 97 and Japan's H-12 were based on mechanistic models. All programmes that have modelled the transport of radionuclides attached to colloids have used a similar one-dimensional transport equation.
- A key uncertainty in assessing the effects of colloid transport is the degree to which the adsorption of radionuclides on colloids is reversible. Most PAs that have considered transport of radionuclides sorbed to colloids assume reversible attachment (e.g. Japan's H-12; Nirex's Nirex 97). The Yucca Mountain TSPA-VA is the only PA in which both irreversible and reversible sorption of radionuclides to colloids was included in PA calculations, and dose comparisons made.
- Five PAs have shown the effects of colloid transport through the geosphere to be potentially significant (NAGRA's Kristallin-I, Japan's H-12, HMIP's Sellafield Assessment, Nirex's Nirex 97, and the US DOE's TSPA-VA for Yucca Mountain). However, in the case of NAGRA's Kristallin-I, the significance was only in the percentage increase of a very small dose and did not jeopardise safety. In the case of HMIP's Sellafield Assessment, the calculations depended critically on assumptions concerning colloid concentrations and the reversibility of radionuclide sorption on colloids, and indicated that colloids could cause a large increase in dose in a small number of simulations. In the US DOE's TSPA-VA, in a small number of simulations there is a one order of magnitude increase in dose in the 50,000 to 250,000 year period (see Section 3.2 for details).

Country, Organisation, PA	Key features (host rock, waste type, hydrological regime)	Colloids included in the FEP list	PAs assuming colloid filtration by bentonite	PAs accounting for colloids in the radionuclide source term	PAs deferring consideration of colloid transport	PAs eliminating geosphere colloid transport	PAs indicating colloid transport is potentially significant
Belgium, ONDRAF/NIRAS	Clay, HLW, Saturated	Yes	No information	No	Yes	-	-
Canada, AECL, EIS	Crystalline, Spent Fuel, Saturated	Yes	Yes	No	No	Yes	-
Finland, POSIVA, TILA-96 TILA-99	Crystalline, Spent Fuel, Saturated	Yes	Yes	No	Yes	-	-
Japan, JNC, H-12	Crystalline, HLW, Saturated	Yes	Yes	Yes	No	No-	Yes
Netherlands, OPLA, PROSA	Salt dome, HLW, Variable	Yes	No	No	Yes	-	-
Sweden, SKI, SITE-94	Crystalline, Spent Fuel, Saturated	Yes	Yes	No	Yes	-	-
Sweden, SKB, SR-95, SR-97 SFL 3-5	Crystalline, Spent Fuel, Saturated	Yes	Yes	No	No	Yes	-
Switzerland, NAGRA, Kristallin-I	Crystalline, HLW, Saturated	Yes	Yes	Yes	No	No	Yes
UK, HMIP, Sellafield Assessment	Crystalline, LLW / ILW, Saturated	Yes	No	Yes	No	No	Yes
UK, Nirex, Nirex 97	Crystalline, LLW / ILW, Saturated	Yes	No	Yes	No	No	Yes
USA, DOE, WIPP	Bedded Salt, TRU, Variable	Yes	No	Yes	No	Yes	-
USA, DOE, Yucca Mountain TSPA-VA	Unsaturated ash-flow tuffs	Yes	No	Yes	No	No	Yes

Table 2.1: *The treatment of colloids in recent PAs, including those reviewed in this report, and those reviewed in Bennett et al. (1999).*

3 Treatment of Colloids in Recent Performance Assessments in Sweden and at Yucca Mountain

In this Section, we provide a summary of the treatment of colloids in recent PAs in Sweden, and in the US DOE's TSPA that was conducted as part of the Yucca Mountain Viability Assessment. Our comments on SKB's treatment of colloids in these PAs are provided in Section 6 of this report. A review of the NTS observations that have been accounted for in the TSPA-VA is provided in Sections 4 and 5 of this report.

3.1 Sweden

SKB has responsibility for disposal of Sweden's radioactive waste. Spent nuclear fuel is an important constituent of the Swedish radioactive waste inventory. At present, spent fuel is stored for about one year at the reactor sites after which time it is transferred to a central interim storage facility. SKB plans that after 30 to 40 years of interim storage, the fuel will be encapsulated in copper canisters and disposed at a depth of about 500 m in crystalline bedrock. This disposal concept is known as KBS-3. The requisite facilities, an encapsulation plant and a deep repository have not yet been sited or built. However, SKB has published several safety assessments that analyse the long-term safety of the KBS-3 system for deep disposal of spent nuclear fuel. In this section we review the way in which colloids have been treated in SKB safety assessments, paying particular attention to the most recent assessment, SR-97. We also review the treatment of colloids in the recently published SFL 3-5 safety assessment of a repository for long-lived low- and intermediate-level wastes.

3.1.1 SKB-91 and SR-95

In 1991, SKB performed a safety assessment of a hypothetical deep repository for spent nuclear fuel at a depth of about 500 m in crystalline basement (SKB-91; SKB, 1992). In 1995, SKB released a template for future safety reports (SR-95; SKB, 1995), which was intended to describe the underlying assessment premises and serve as a platform for future safety assessments. Screening calculations for processes related to colloids were performed to support the 1991 PA (Allard *et al.*, 1991). These calculations were updated, and alternative scenarios explored, as part of the 1995 safety report (Laaksoharju *et al.*, 1995).

The following colloid-related FEPs were included in SKB's list of potentially relevant features, events and processes (FEPs) to consider in their PAs (Andersson, 1989):

- 3.1.4 Colloid generation - source
- 3.2.4 Erosion of buffer/backfill
- 4.1.3 Colloids, complexing agents
- 4.1.9 Complexing agents
- 5.45 Colloid generation and transport

Screening

All of the above FEPs were included within the reference (normal evolution) scenario for the 1991 PA, with the exception of FEP 4.1.3, which was cross-referred to FEPs 4.1.9 and 5.45.

The concentrations of inorganic colloids in Swedish crystalline basement groundwaters were measured as a function of depth by Laaksoharju (1990). Deep Swedish groundwaters tend to be characterised by high ionic strength and high concentrations of divalent ions such as Ca^{2+} . These properties tend to destabilise colloids, and this was reflected in the low measured colloid concentrations observed. The composition of the colloids varied with depth, and calcite particles dominated in the deeper samples. Also present at lower abundance were iron hydroxide, gibbsite, iron sulphides and quartz particles.

The screening calculations performed by Allard *et al.* (1991) used a mean total concentration value of $10^{-4} \text{ kg m}^{-3}$, a minimum value of $10^{-5} \text{ kg m}^{-3}$ and a maximum value of $4 \times 10^{-4} \text{ kg m}^{-3}$. The measured concentrations on which these values were based were re-evaluated by Laaksoharju *et al.* (1995), who noted that calcite and possibly sulphide colloid concentrations were largely an artefact of the sampling and measurement procedures. Omitting these colloids resulted in much smaller calculated colloid concentrations of 2 to $4.5 \times 10^{-5} \text{ kg m}^{-3}$, with the higher concentrations in the less saline, shallower groundwaters.

The concentration of microbes was measured at depths ranging from 100 to 900 m at various sites in Sweden using a downhole probe (Pedersen, 1990). Typical values were around 100,000 per ml. For the screening calculations by Allard *et al.* (1991), this was converted to a mean value of $10^{-5} \text{ kg m}^{-3}$, assuming a mass of 10^{-16} kg per cell, and a maximum of $5 \times 10^{-5} \text{ kg m}^{-3}$. The concentration of humic and fulvic acids was estimated based on measurements of total organic carbon, giving a mean value of $10^{-4} \text{ kg m}^{-3}$ and a maximum of $5 \times 10^{-4} \text{ kg m}^{-3}$.

Allard *et al.* (1991) reported screening calculations for the effects of inorganic colloids, microbes, and humic and fulvic acids on radionuclide transport. High and low ionic strength (chloride concentration = 5500 and 61 mg l^{-1} , respectively), high and low pH (8 and 6, respectively), and the presence or absence of humics were taken into account. Distribution coefficients were derived describing sorption of key radionuclides onto the granitic rock and onto the inorganic and microbial colloids for a reference case (neutral pH and reducing conditions in the presence of humic substances; see Table 3.1). Experimental investigations of sorption onto inorganic particles (Sätmark and Albinsson,

1990) and onto microbes (Pedersen and Albinsson, 1990) were used as a basis for the derivation.

Assuming the sorption of radionuclides to colloids to be reversible, and neglecting colloid filtration and channelling, the effect of colloids on radionuclide transport was examined by Allard *et al.* (1991). An effective distribution coefficient, K_{eff} , was defined:

$$K_{\text{eff}} = K_d^{\text{rock}} / (1 + C \cdot K_d^{\text{particle}}) \quad (1)$$

where:

C = the particle (inorganic colloids and/or microbes) concentration.
 K_d^{particle} = the distribution coefficient for sorption onto the particle.

For the combination of the highest colloid concentrations ($5 \times 10^{-4} \text{ kg m}^{-3}$) and highest K_d^{particle} ($30 \text{ m}^3 \text{ kg}^{-1}$), the effective distribution coefficient is only reduced from K_d^{rock} by 1%. As this effect is negligible compared to the uncertainty associated with the K_d^{rock} values, the effects of reversible sorption onto colloids were considered to be of low significance to the PA calculations.

The effects of irreversible radionuclide sorption on colloids were calculated by assuming that a fraction of the radionuclides become irreversibly bound to colloids as they leave the near-field, and that they then migrate at the same speed as the groundwater. This represents a conservative approach compared to the assumption in SKB's PAs that all repository-derived colloids will be filtered by the bentonite surrounding the canisters (see Section 2).

The fraction of the radionuclides released from the near-field that are bound to colloids was calculated from:

$$C \cdot K_d^{\text{particle}} / (1 + C \cdot K_d^{\text{particle}}) \quad (2)$$

giving a maximum value of 0.003 at the highest observed colloid concentration. Allard *et al.* (1991) used estimates of the release of uranium from the near-field of the repository to make dose estimates for radionuclides carried by colloids. It was assumed that the repository contained 6,000 canisters, each containing one tonne of uranium. After 1,000 years, six canisters were breached and were leached at a rate of 10^{-7} per year. The fraction of radionuclides attached to colloids was calculated from Equation (1). All radionuclides attached to colloids were assumed to flow to a well at a total flow-rate of $4,000 \text{ m}^3$ per year. A person was assumed to drink 600 litres of water per year from the well. Because of the small carrying capacity of colloids, Allard *et al.* (1991) found that the potential dose from the colloid fraction reaching the drinking water well was less than 1 mSv per year, even for a highly conservative scenario in which all colloids released from the repository go directly to a drinking water well.

Oxidation State	Element	$K_d(\text{rock})$	K_{part} Colloids	K_{part} Bacteria
M(I)	Cs	0.03 ^f	0.01	0.01
M(II)	Sr	0.003 ^f	1	0.01
	Ra	0.03 ^f	10	0.1
	Ni	0.006 ^f	1	0.02
	Pb	0.3	10	0.02
	Pd,Sn	0.001	0.001	0.001
M(III)	Ln,An ^b	0.2	30	0.01
M(IV)	An ^c	2	30	0.5
	Tc,Zr,	1	30	0.2
	Nb ^d ,Pa	1	30	0.2
Others	C	0.001	0.001	25
	X ^e	0	0	0
	Se	0.001	0.001	0.001

^b Lanthanides; Ce, Sm; actinides: Pu, Am

^c Actinides; Th, U, Np

^d Nb is pentavalent

^e Halogens; Cl, I

^f For LIS these values are increased by a factor of 5

Table 3.1: Distribution coefficients ($m^3 \text{ kg}^{-1}$) for selected elements onto rock, inorganic colloids and organic particulates (bacteria) under reference chemical conditions (high ionic strength, neutral pH, reducing conditions, presence of humic substances, low radionuclide concentrations). The data are taken from Allard et al. (1991).

The occurrence of chemical and physical processes that could change the colloid population and/or change colloid migration behaviour was evaluated by Laaksoharju *et al.* (1995). Chemical changes, such as a decrease in salinity due to an influx of fresh glacial meltwater during deglaciation, were considered to be operative only over a short timescale. Although a decrease in salinity could increase colloid stability, changes in colloid concentrations in excess of an order of magnitude above the current background level were not anticipated. Furthermore, dilution of the deep saline groundwater was calculated to be less than 50%, and the Ca concentrations at the near-field/far-field interface were expected to remain above 10^{-4} molar.

Transport of radionuclide-bearing colloids by gas bubbles was considered to merit further study, following the work of Wan and Wilson (1994a,b) and Neretnieks and Ernstsson (1997).

3.1.2 SR-97

SKB's SR-97 safety assessment attempts to demonstrate that the KBS-3 concept has good prospects of being able to meet the radiation protection requirements specified by SKI and SSI (SKB, 1999a). SR-97 considers three different sites, characteristic of three different Swedish granitic bedrock environments. The assessment applies to a closed repository for spent nuclear fuel.

The future evolution of the repository is analysed in the form of five scenarios. The first is a base scenario where it is postulated that the repository is built according to specifications, and present-day environmental conditions are postulated to persist indefinitely. The four other scenarios illustrate the evolution of the repository for situations that differ from the base scenario:

- The repository contains a few initially defective canisters.
- Significant climate change occurs.
- Significant seismicity occurs.
- Future inadvertent human intrusion occurs.

In the SR-97 safety assessment, colloids are screened out of all scenarios because SKB considers it unlikely that they will be present in significant quantity. In the base scenario, extensive colloid formation is expected to occur temporarily, in connection with the closure of the repository. Within a few years, the concentration is expected to fall to less than 0.5 mg per litre (Laaksoharju *et al.*, 1998), and colloids are therefore considered by SKB to be of low consequence to the performance of the disposal system. Similarly, in the canister defect scenario, the expected colloid concentrations are expected to be of low consequence to performance of the disposal system owing to filtration by the bentonite.

Repository evolution is divided into thermal, hydraulic, mechanical and chemical (THMC) processes, and the analyses are intended to evaluate the capacity of the repository to isolate the radioactive waste in the canisters, or to retard any releases of radionuclides if the canisters are damaged. The timescale for the analyses is at most one million years, in accordance with preliminary regulatory requirements.

In SR-97, colloids are considered under the category of chemical processes. THMC influence diagrams constructed for the spent fuel, buffer and backfill, and for the geosphere, consider processes related to colloids. The diagrams identify processes acting on a particular part of the repository, and list the main variables that influence each process, as summarised below:

Spent Fuel

Process

Speciation of radionuclides/colloid formation

Variables affecting process

Temperature

Radiation intensity

Material composition

Water composition

Radionuclides in the fuel matrix may be released in the form of colloids or may form pseudocolloids. SKB considers that as long as the bentonite buffer remains in place, such colloids are of no importance for radionuclide dispersal from the canister, because the buffer will act as an efficient filter of colloids (Kurosawa *et al.*, 1997).

Leaching experiments were performed by SKB in order to determine the speciation of radionuclides derived from spent fuel under typical repository conditions (Forsyth, 1997). Filtration experiments were carried out on the leachates produced in these experiments in order to determine the significance of colloids. However, only one filter was used to filter the leachates with an aperture of 15-20 μm . The material on the filter ranged from 15-20 μm to hundreds of microns in size, and was probably dominated by particles larger than colloids. The concentration of uranium in the filtered material was about 0.1 ppm, as compared with a few ppm for the uranium in solution under oxidising conditions.

Considerable uncertainty remains in the potential for colloid generation from corrosion of spent fuel in the repository. However, it is assumed that the buffer will filter all colloids that may be produced, and the process is therefore screened out of the SR-97 assessment.

Buffer and Backfill

Process

Radionuclide transport by colloids:

Variables affecting process

Temperature
Water content
Hydrological parameters
Buffer geometry
Pore geometry
Smectite composition
Pore water composition

Colloid release/erosion process:

Hydrological parameters
Smectite composition
Pore water composition
Smectite content

SKB considers that colloids could form during the dissolution of the spent fuel. Diffusive transport of colloids through highly compacted bentonite is assumed to be negligible, due to the tortuosity and small size of the bentonite pores. SKB cites as an example experiments on 15-nm gold colloids that demonstrate that bentonite with a dry density of $1,000 \text{ kg m}^{-3}$ effectively filters colloids (Kurosawa *et al.*, 1997).

Colloid release/erosion processes involve release of colloids from the clay buffer. During expansion, bentonite from the buffer may intrude fractures around the deposition hole and fill voids in the repository tunnels. Groundwater can erode the buffer as a result of:

- Mechanical erosion where particles are torn loose by the flowing water.
- Chemical erosion where low salinity groundwater disperses the clay gel.

The sensitivity of the buffer to erosion depends on its density, which in turn depends on the external pressure to resist swelling. If there is no resistance, the bentonite may swell to a low-density gel. In a fracture, the resistance is close to zero at the point of contact between the bentonite and the water. In the repository, the risk of erosion is greatest if there is a gap between the backfill and the roof of the tunnel.

Erosion and release of colloids is dependent on groundwater composition and flow rates:

- The ionic strength of the groundwater, particularly the concentration of the more highly charged, highly polarising cations, determines whether the expanded clay gel will dissolve. If the concentration of divalent ions such as Ca^{2+} is greater than about 4 ppm, the clay gel will be stable. This is the case in most deep Swedish groundwaters (Allard *et al.*, 1991). A high concentration of univalent ions such as Na^+ will also stabilise expanded clay gels, but in this case the ionic strength must be much higher.

- If the groundwater flow velocity is sufficiently rapid, the outermost layer of clay may be eroded as colloid-sized particles. A water velocity of at least 1,000 times the mean water flow velocity in the water-saturated repository is required in order for erosion of expanded clay gels in fractures to occur (Le Bell, 1978). This implies that under normal hydraulic gradients, erosion of bentonite will not occur.

Experiments carried out within the European Commission CARESS project provided constraints on the measured concentration of colloids generated from bentonite in contact with granitic groundwater (Gardiner, 1997). These data were cited by SKB as evidence that colloid formation would be negligible. The experiments showed that with a saturated buffer, the leached concentration of colloids was below 1 ppm.

SKB concluded that erosion of the buffer and formation of colloids is not of significance to the long-term performance of the repository. The process was therefore screened out of the SR-97 safety assessment, but SKB considers that it should be further studied.

SKB has recently published a report evaluating colloid generation by erosion of bentonite buffer clay that has penetrated into fractures (Pusch, 1999). The report considers that most colloids formed will be immobilised by sorption to chlorite and other mineral fracture fillings, and that colloid formation will be limited to the first few years after buffer emplacement because it is only at this time that flow rates are sufficiently high to erode the buffer clay. The report concludes that the chance of such colloids reaching the biosphere is very low.

Geosphere

Process

Colloid formation:

Variables affecting process

Temperature
Groundwater flow
Fracture minerals
Groundwater composition

Radionuclide transport by colloids:

Temperature
Groundwater flow
Repository geometry
Fracture flow
Matrix minerals
Fracture minerals
Groundwater composition

SKB believes that colloidal particles can form in the groundwater as a result of chemical reactions, and as a result of erosion. Radionuclides may become attached to such colloids and be transported by them.

In general, the concentration of colloids in deep Swedish groundwaters is quite low. According to Allard *et al.* (1991) and Laaksoharju *et al.* (1993), the concentration is less than 0.5 mg per litre. This value has been revised to even lower levels by Laaksoharju *et al.* (1995), who considered that the calcite-bearing and sulphur-bearing colloids were

artefacts. The revised colloid concentration is reported by Laaksoharju *et al.* (1995) to be in the range 20 to 45 µg per litre (ppb). However, this value is not cited in SR-97 and SKB simply states that concentrations of colloids are likely to be below 0.5 mg per litre (TR-99-07, pages 242 and 257).

SKB therefore believes that colloids are not generated in significant quantities under these chemical conditions. In contrast, mixing of atmospheric oxygen with a reducing groundwater tends to promote extensive colloid formation. It is therefore likely that after closure of the repository, the water may contain high concentrations of colloidal material, of the order tens of ppm, during a transitional period between closure and the re-establishment of normal background chemical conditions (Wikberg *et al.*, 1987). SKB expects the transitional period to last a few years.

SKB recognises two end-member situations in the attachment of radionuclides to colloids, reversible attachment and irreversible attachment:

- Reversible attachment will result in a reduction of the K_d value for sorption on the host rock of a given radionuclide. This reduction is considered negligible given the maximum colloid concentration observed in deep Swedish groundwaters (Allard *et al.*, 1991; Laaksoharju *et al.*, 1995).
- Irreversible attachment may in the most extreme case result in a radionuclide being transported without any retardation due to sorption or matrix diffusion. However, SKB believes that even in this case, the colloid concentrations are so low that the consequences for safety are negligible, based on scoping calculations carried out by Allard *et al.* (1991).

In summary, SKB believes that although processes connected with colloids are subject to considerable uncertainty, the concentration of colloidal material that is required in order to have a significant impact on repository performance will never occur under natural conditions in the groundwaters in question.

3.1.3 SFL 3-5

Low- and intermediate-level waste (LLW and ILW) from the operation of Swedish nuclear power plants, and smaller amounts from medical, research and industrial sources, is treated, packaged and shipped to the SFR repository in Forsmark. Some of the LLW and ILW contains too much long-lived radioactive material for disposal at SFR. SKB is therefore planning the disposal of long-lived LLW and ILW in a deep repository, SFL 3-5.

SKB performed a preliminary safety assessment for the deep disposal of Swedish LLW and ILW in the SFL 3-5 repository (SKB, 1999c). The purpose of the study was to investigate the capacity of the facility to act as a barrier for the release of radionuclides, and toxic pollutants such as lead, beryllium and cadmium, and to assess the importance of the location of the repository site. It was assumed that the SFL 3-5 repository was co-

sited with the SR-97 deep repository for spent fuel. The SFL 3-5 assessment therefore assumed the same three hypothetical sites that were assumed in the SR-97 PA.

The study focused on quantitative evaluation of the reference scenario. The reference scenario described the expected evolution of the near-field of the repository, assuming stable hydrological, chemical, thermal and mechanical conditions in the rock surrounding the repository. Other scenarios were discussed and analysed in more general terms. It was found that mobile, long-lived radionuclides such as ^{36}Cl and ^{93}Mo were most important to the safety assessment. Two factors were identified as being of particular importance:

- The water flow at the depth at which the repository is built.
- The ecosystem at the ground surface in areas where future releases may take place.

Colloids were considered as a possible means of transport for radionuclides. However, by reference to the study of Allard *et al.* (1991), SKB states that the capacity of colloids to sorb and transport radionuclides is small compared with the rock's capacity to sorb and retard transport. Transport of radionuclides by colloids was excluded from the safety assessment because the quantity of colloids was expected to be very small.

3.2 United States: Yucca Mountain Viability Assessment

The United States Department of Energy (DOE) has been studying a site at Yucca Mountain, Nevada, for more than 16 years to determine whether it is a suitable place to build a geological repository for US commercial and defence spent nuclear fuel and high-level radioactive waste (HLW). In the 1997 Appropriations Act, the US Congress required the DOE to prepare an assessment of the viability of the Yucca Mountain site. In order to satisfy this requirement, in December 1998 the US DOE published a Viability Assessment of a repository at Yucca Mountain (US Department of Energy, 1998). The Viability Assessment (VA) provided information on the progress of the Yucca Mountain Site Characterisation Project and comprised:

- The preliminary design concept for the critical elements of the repository and waste package.
- A Total System Performance Assessment (TSPA) of a future Yucca Mountain repository.
- A plan and cost estimate of the remaining work required to complete and submit a licence application to the Nuclear Regulatory Commission (NRC).
- An estimate of the costs to construct and operate the repository.

The TSPA-VA used all available scientific data and analysis in conducting a PA of the Yucca Mountain system, and included explicit consideration of the influence of colloids on radionuclide transport. The PA and sensitivity studies were intended to give a broad indication of the probable system behaviour. However, the assessment was not as detailed as a future licence application might be expected to be. For example, the TSPA-VA did not include a comprehensive FEP screening analysis.

3.2.1 Yucca Mountain Design Concept

The design concept for the Yucca Mountain repository system involves the emplacement of various waste forms, including spent fuel and vitrified HLW, within packages comprising an inner layer of corrosion-resistant nickel alloy, and an outer layer of carbon steel. The waste packages are placed on supports within mined drifts located about 300 m below the surface but above the water table.

The TSPA-VA considered the migration of rainwater from the ground surface through the unsaturated zone, and the interaction of this water with the waste in the repository. The downward migration of radionuclides released from the waste form, to the water table, was also modelled, together with transport in the underlying regional hydrological system. Radionuclides released from the repository were modelled as entering the biosphere in populated regions, several tens of kilometres to the south, via a well pathway. The evolution of the near-field environment within the drifts, and interaction of the waste with groundwater, were represented using coupled process models.

The TSPA-VA indicated that most of the radionuclides in the waste are essentially immobile and never leave the repository. A few radionuclides, including ^{99}Tc , ^{129}I , ^{237}Np , and those radionuclides transported by colloids, are sufficiently mobile that they could reach the biosphere downgradient from the repository.

3.2.2 Treatment of Colloids

Recent observations of Pu, and other radionuclides, associated with colloids at the Nevada Test Site (NTS), adjacent to Yucca Mountain, suggested that colloidal transport of Pu should be considered in the Yucca Mountain performance assessment (Thompson, 1998; Kersting *et al.*, 1999). Pu is a major constituent of the Yucca Mountain inventory, has low solubility and high sorption onto the host rock, and is the radionuclide most likely to be affected by colloidal transport. Colloid-assisted transport of Pu was therefore included within the TSPA-VA. A detailed description of the treatment of colloids is provided in one of the underlying supporting documents to the TSPA-VA (CRWMS M&O, 1998). Colloidal transport of other radionuclides was not considered in the TSPA-VA but will be investigated by the Yucca Mountain programme in the future as more data become available.

Near-Field

Many types of colloid are expected to be present in the Yucca Mountain repository system, but only four types were chosen for explicit treatment within the near-field model: clay, iron corrosion products, colloids produced by degradation of spent nuclear fuel, and colloids produced by degradation of vitrified waste. These four colloid types were chosen because they may be present at potentially significant concentrations.

A set of five near-field models was used to represent the near-field geochemical environment. These models described:

- Incoming gas, water and colloids.
- The gas phase within the drifts containing the waste packages.
- The chemistry of the water and gas phases within the drifts.
- The stability and quantity of colloids within the drifts.
- The microbial communities within the drifts.

The first and third of these models dealt with the behaviour of colloids in the Yucca Mountain system. The interrelations between the five near-field models are shown in Figure 3.1. In some cases, there are a number of sub-models to represent detailed processes within the main near-field models.

The flux of water and dissolved constituents through the unsaturated zone surrounding the repository is given either as a seepage flux through the drifts, or as an average percolation flux at the repository horizon. Seepage flux is used for the in-drift water model and the colloid model.

Incoming gas, water and colloids

The near-field geochemical environment model provides values for the time-varying compositions and fluxes of gas, water and colloids entering the drift. These are used as time-dependent boundary conditions for the models of chemical reactions in the drift. Changes to these variables are primarily driven by thermal changes to the system. Reactions between gas, minerals and water will occur after repository closure, as the system is first heated and pore fluid boils, then subsequently cools with accompanying condensation of water vapour.

The model for incoming colloids assumes that colloids entering the drift are the same as those found in the natural far-field. The incoming colloids model includes an estimate of the types of colloids that may enter the drift, and general constraints on the amounts of those colloid types. Typical naturally occurring colloids in Yucca Mountain groundwaters are composed of clays, silica and iron oxyhydroxides at a concentration of about 20 to 30 ng per ml (Triay *et al.*, 1996). Because the iron-oxyhydroxide colloids generated within the drift represent a larger potential source than the incoming natural iron-oxyhydroxide colloids, the incoming colloids are represented by the clay component of the natural colloids.

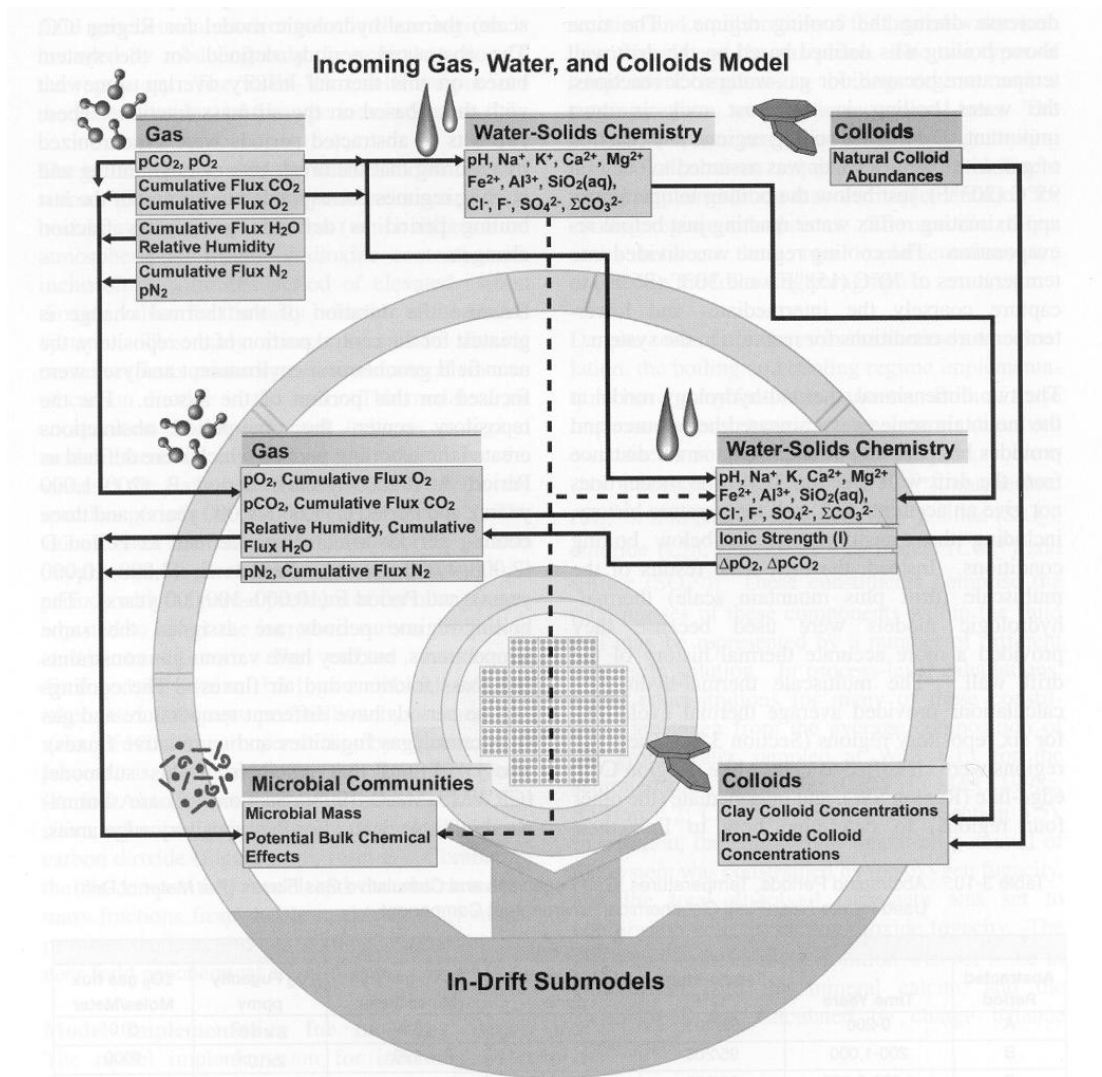


Figure 3.1: Flow diagram depicting the interrelations and parameter exchanges among all near-field geochemical environment models in the TSPA-VA (after US Department of Energy, 1998).

The amounts of clay and iron oxyhydroxide colloids in the Yucca Mountain repository are constrained by the ionic strength of the fluid in the drift. Measurements of the naturally occurring abundance of colloids in Yucca Mountain groundwaters were used to demonstrate that the relationship between colloid abundance and ionic strength at Yucca Mountain is similar to that observed in other groundwater systems worldwide, for colloids greater than 100 nm in size (Triay *et al.*, 1996) (see Figure 3.2).

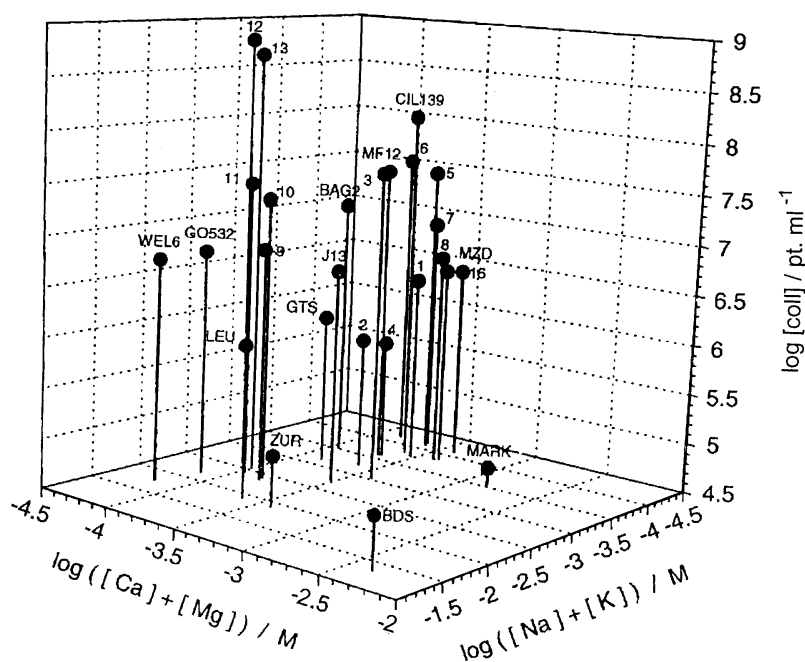


Figure 3.2: Relationship between colloid concentration, salt concentration and water hardness for groundwaters sampled from a variety of locations worldwide, after Triay et al. (1996) and CRWMS M&O (1998).

Although colloids smaller than 100 nm exist, the data available to constrain colloid abundance is most accurate for colloids greater than this size. The relationship may therefore not account for those smaller colloids that potentially could account for about 10-40% of colloidally transported radionuclides. The colloid abundance relation and its associated uncertainty were implemented directly in the models of the near-field geochemical environment.

The TSPA-VA divides the evolution of the repository into seven periods:

- Time of waste emplacement to time of repository closure.
- Time of repository closure to several hundred years after closure.
- Several hundred years to several thousand years after closure.
- Several thousand years to ten thousand years after closure.
- Ten thousand years to several tens of thousands of years after closure.
- Several tens of thousands of years to one hundred thousand years after closure.
- Several hundred thousand years to one million years after closure.

For each period, the calculated ionic strength of near-field fluids was used, together with the relationship linking colloid concentration to ionic strength, to assess the stability and concentration of the suspended colloids reacting with the waste form. The ionic-strength/colloid stability relationship was used, external to the system code, to estimate amounts of clay and iron-oxyhydroxide colloids in the drift, for different ionic strengths.

Under typical boiling regime conditions with an ionic strength of about 3×10^{-2} molal, a colloid concentration of about 8×10^{-6} mg/ml, or 1×10^6 particles per ml, was estimated. Under typical cooling regime or ambient conditions, with an ionic strength of about 5×10^{-3} molal, a colloid concentration of about 6×10^{-5} mg/ml, or 7×10^6 particles per ml, was estimated. These parameters were used as input to the colloid sorption and transport model for Pu concentrations.

Far-Field

Colloids may not be able to move through the rock matrix, particularly the welded and zeolitic rock types, because of colloid size relative to matrix pore size. However, movement through the more permeable, unwelded vitric rock of the Calico Hills tuff may be possible. In this higher permeability unit, advective aqueous phase transport of radionuclides between fractures and matrix was assumed to characterise the flow field. As an approximation to simplify the transport model, colloids were also allowed to move by advection with the aqueous phase radionuclides, between the fractures and the matrix. However, colloids were not allowed to exchange between fractures and matrix through matrix diffusion. Colloid transport was assumed to be unretarded through the fracture continuum.

The transport of aqueous and colloidal radionuclides passing through fractures was expressed in terms of advection, sorption onto fracture surfaces, matrix diffusion, dispersion and radioactive decay. Additional equations were used to describe the advection, sorption, dispersion and decay of radionuclides attached to colloids. Colloid particles themselves were assumed not to experience any significant matrix diffusion because of the very low diffusion coefficients associated with colloids.

Mathematical Representation

In the TSPA-VA, it was assumed that sorption of radionuclides on colloids may be approximated using a K_d model. No distinction was made between different colloid types. Advective transport through the fractures was enhanced by reduced matrix diffusion and sorption. By assuming a constant concentration of colloids, and by applying the K_d model for sorption, the ratio of the radionuclide mass in colloidal form to that in aqueous form was calculated to be a constant value (K_c).

Reversible, linear sorption of contaminants onto colloids was assumed:

$$C_c = K_c C \quad (3)$$

where:

- K_c = the aqueous-colloid partitioning coefficient
 K_c = a dimensionless parameter expressing the ratio of contaminant mass residing on the colloids to the mass present in aqueous form.

K_c is also the product of the distribution coefficient for contaminant on colloids and the concentration of colloidal material available for sorption.

The mathematical transport model for aqueous and colloidal radionuclides was thus reduced to two simple equations in terms of the aqueous radionuclide concentrations. The colloid transport equation has the same structure as that for aqueous transport, but also includes information concerning colloid partitioning.

The equation for aqueous transport alone considers 1-D advection-dispersion, diffusion into the porous rock matrix, equilibrium, and linear sorption onto the fracture surface:

$$R_f \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - n \frac{\partial C}{\partial z} - \frac{2q}{b} \quad (4)$$

where:

- C = concentration [kg/m³]
 t = time [s]
 z = the coordinate in the direction of fluid flow [m]
 R_f = the sorption retardation factor for the contaminant on the fracture
 D_z = the axial dispersion coefficient [m²/s]
 m = the fluid velocity [m/s]
 q = the diffusive loss term for the porous matrix [m²/s]
 b = the fracture aperture [m]

An equation for transport of a contaminant attached to colloids may be written as:

$$R_c \frac{\partial C_c}{\partial t} = D_z \frac{\partial^2 C_c}{\partial z^2} - n \frac{\partial C_c}{\partial z} \quad (5)$$

where:

- C_c = the concentration of contaminant attached to colloids [kg/m³]
 R_c = the retardation factor that captures colloid attachment and detachment to and from the rock, and colloid filtration processes.

Reversibility of Colloid Attachment

The reversibility of Pu attachment has been investigated experimentally by the US DOE (CRWMS M&O, 1998). Studies of Pu attachment on hematite, goethite and clay show fast attachment and slow detachment. The DOE expects sorption of dissolved Pu onto the major near-field and far-field colloids to be reversible on the time scale of transport to the accessible environment (hundreds of thousands to millions of years). However,

co-precipitation may create colloids in which radionuclides are encapsulated by stable minerals. Such radionuclides would effectively be irreversibly attached to the colloids.

In the TSPA-VA, Pu attachment to colloids was modelled using two extremes: instantaneous reversible attachment, and totally irreversible attachment, as illustrated in Figure 3.3.

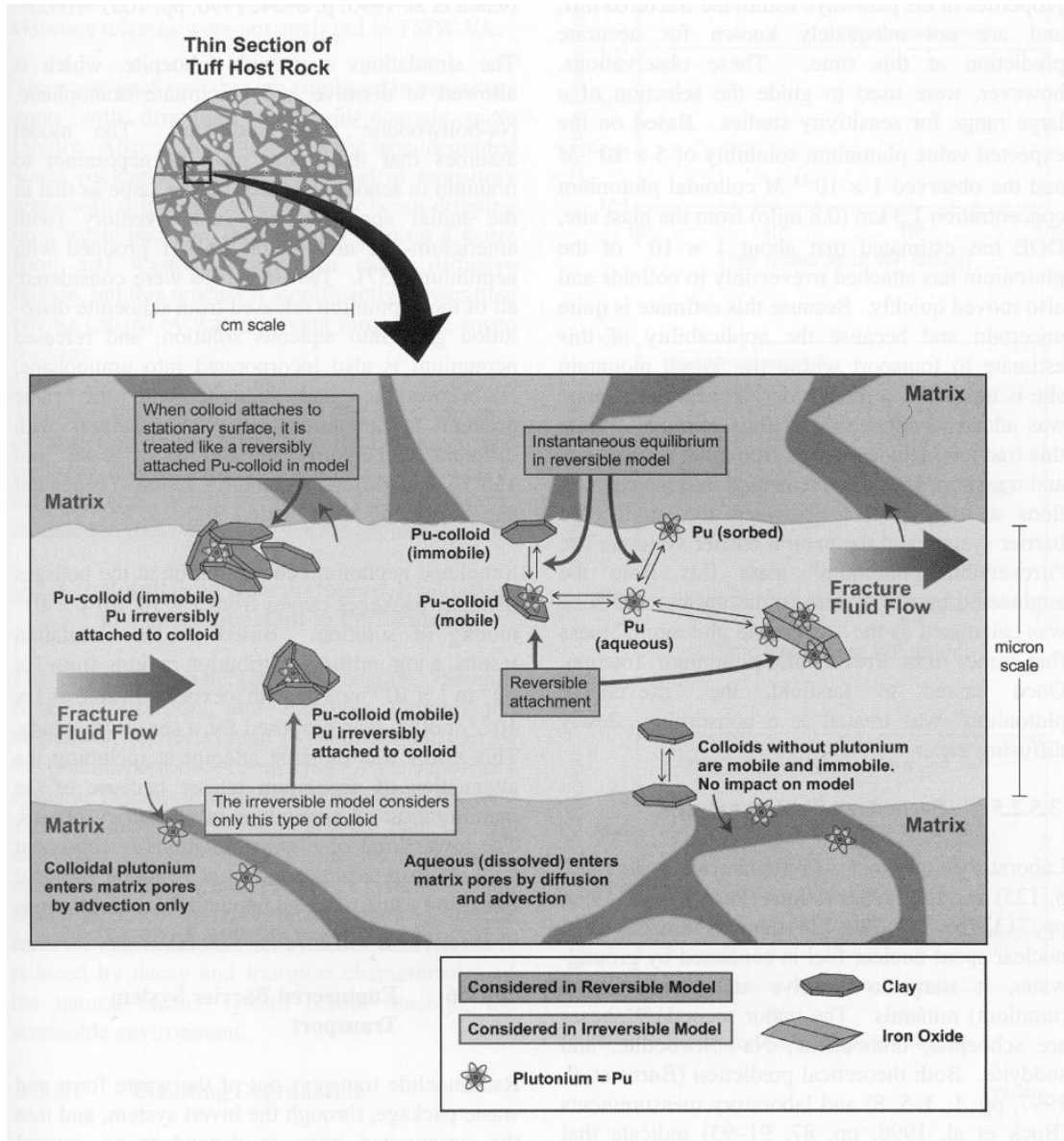


Figure 3.3: Illustration of two models for colloidal transport of plutonium, involving reversible plutonium attachment and irreversible plutonium attachment, used within the TSPA-VA (after US Department of Energy, 1998).

Coupling the transport equation for contaminants attached to colloids, Equation (5), with the aqueous transport equation, Equation (4), and using the expression for C_c given in Equation (3), the following equation is obtained:

$$\left(\frac{R_f + K_c R_c}{1 + K_c} \right) \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - n \frac{\partial C}{\partial z} - \frac{2q}{b(1 + K_c)} \quad (6)$$

The coupling between fracture and matrix is identical to the case without colloids because no migration of colloids into the rock matrix is assumed. In order to allow for colloids, the parameter R_f in Equation (4) is replaced by:

$$\hat{R}_f = \frac{R_f + K_c R_c}{1 + K_c} \quad (7)$$

representing a combined retardation factor for colloids and dissolved contaminants.

Reversible Model

The reversible model assumes instantaneous equilibrium between dissolved radionuclides and colloids, and was implemented within the RIP code for use in the near-field, and the FEHM code for use in the far-field. The RIP model defines the amount of radionuclide mobilised in colloidal form as the product of the dissolved radionuclide concentration, the dissolved radionuclide-colloid sorption partition coefficient (K_d), and the colloid concentration. The colloid concentration was calculated as a function of the ionic strength of the solution as described above.

The FEHM model combines the K_d and colloid concentration parameters into a single parameter, the aqueous-colloid partitioning coefficient, K_c . The parameter ranges used in the calculations are summarised in Table 3.2.

	K_d (mg per g)	Effective K_c
Near-field colloids		
Clay	$10^3 - 10^5$	$10^{-5} - 10^{-2}$
Iron oxyhydroxide colloids	$10^4 - 10^6$	$10^{-4} - 10^{-1}$
Spent fuel waste form colloids	$10^2 - 10^8$	$10^{-5} - 10^0$
Glass waste form colloids	$10^2 - 10^9$	$10^{-5} - 10^1$
Far-field colloids		$10^{-5} - 10^1$

Table 3.2: Parameters used in reversible colloid attachment model.

Irreversible Model

The irreversible model was also included in the TSPA-VA calculations to capture the possibility that a fraction of the radionuclides might move through the system irreversibly attached to colloids. The model was developed in response to the suggestion that irreversible colloid attachment is a possible explanation for the observations at the NTS (see Section 4 of this report and Thompson (1998) and Kersting *et al.* (1999)). Specifics of colloid properties such as surface charge, size distribution, expected stability, and transport and filtration properties within the fractured tuff are not adequately known at present, and so sensitivity studies were made using a wide range of parameter values.

At the NTS, groundwaters 1.3 km from the BENHAM test site contain colloid-associated Pu that can be shown to have been derived from the BENHAM test (see Section 4). The colloidal Pu concentration is 1×10^{-14} M. Based on an assumed Pu solubility of 5×10^{-7} M, the US DOE has estimated that a fraction of about 1×10^{-7} of the Pu may have become attached irreversibly to colloids and travelled 1.3 km in 30 years (Thompson, 1998). Because this estimate is uncertain, and because the applicability of this estimate to transport within the Yucca Mountain site is unknown, a three order of magnitude range was added to either side of this estimate. The “irreversible Pu” mass flux from the engineered barrier system to the unsaturated zone was calculated as the “reversible Pu” mass flux multiplied by the irreversible Pu fraction. Once passed to the far-field, the “irreversible Pu” was treated as a nonsorbing, slowly diffusing tracer.

In the irreversible colloid model, interaction with the aqueous phase and immobile rock phase is neglected. This is accounted for by multiplying Equation (6) by K_c and then letting $K_c \tau \equiv$. Equation (6) then becomes:

$$R_c \frac{\partial C_c}{\partial t} = D_z \frac{\partial^2 C_c}{\partial z^2} - n \frac{\partial C_c}{\partial z} \quad (8)$$

Therefore, colloid transport with irreversible sorption is mathematically analogous to aqueous radionuclide transport with no matrix diffusion and no sorption.

Sensitivity Analysis

Uncertainty in colloid formation and transport parameters, particularly in the far-field transport parameters, is high, because of a lack of experimental or field-scale evidence to corroborate the models. Nevertheless, colloid model parameters do not show up as the most influential in the TSPA-VA consequence calculations.

In the TSPA-VA, the average contribution of each radionuclide to the peak dose rate over three different simulation periods was calculated. The periods were zero to 10,000 years, zero to 100,000 years, and zero to 1,000,000 years. For a given period, the relative individual radionuclide contributions to the peak dose rate for each realisation were calculated, and the relative contribution of each radionuclide was averaged across all realisations. Recalling that Pu was the only radionuclide affected by colloid transport, the Pu dose rate is the most significant contributor to the total peak dose rate for only

2% of the zero to 100,000 year multiple realisations and 8% of the zero to 1,000,000 year realisations. However, in examining the sensitivity to the 95th percentile value of the K_c range ($K_c = 5.0$) in the base case, where K_c is the aqueous-colloid partitioning coefficient, there is a one order of magnitude increase in dose in the 50,000 to 250,000 year period (Figure 3.4).

Model for In-Drift Microbial Communities

A model for in-drift microbial communities was used to provide a bounding assessment on the masses of produced microbes within the disposal system. The model is implemented with a code for microbial impacts to the near-field geochemistry and uses an idealised approach to assess biomass production. At each time step, the analysis determines whether biomass production is limited based on either the available energy in the system or the nutrient supply for microbes: the microbial production for that time step is the lower of the two values. The production rate and the cumulative biomass can then be used to determine the biomass yield. The analyses were not coupled to the colloids model, or used directly within the TSPA calculations, but were used to assess the need for potential future work in this area.

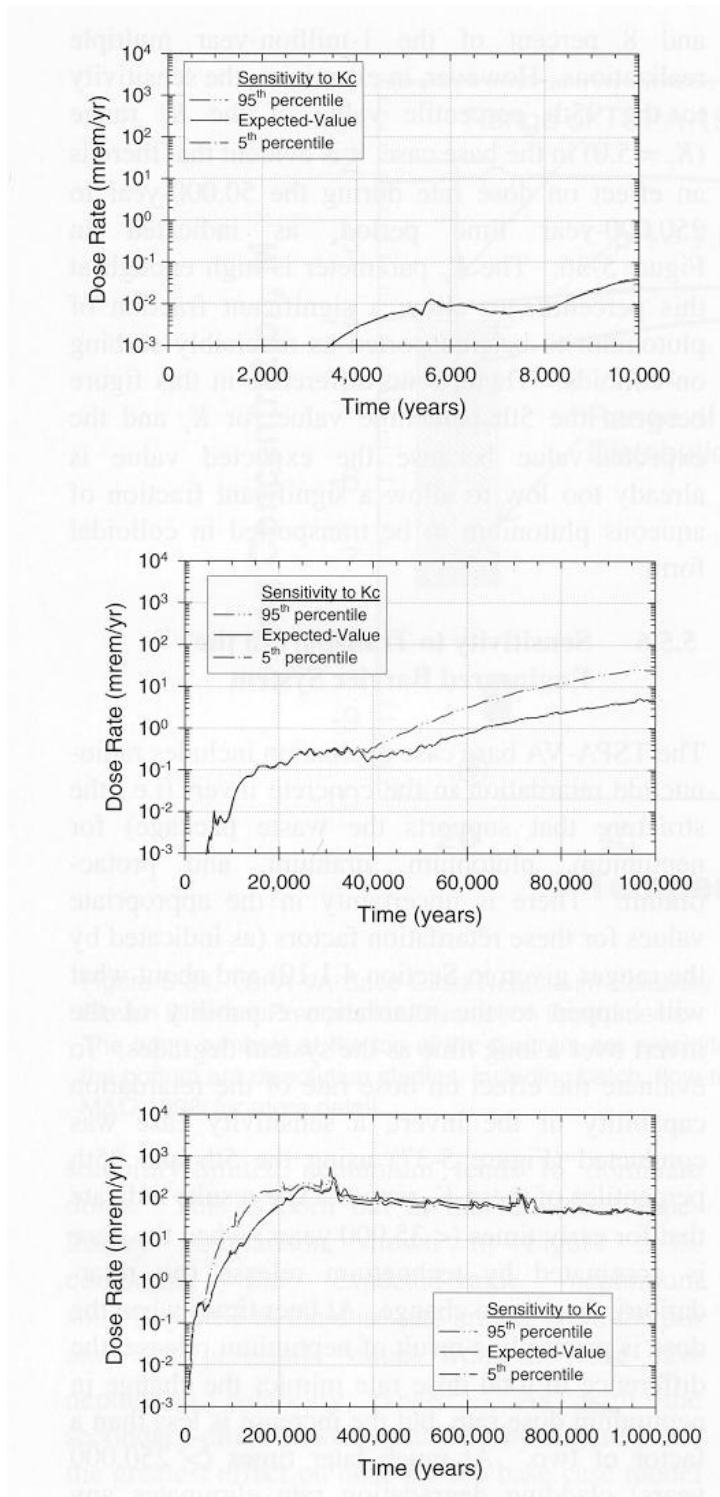


Figure 3.4: *Effect of colloid distribution coefficient on total dose rate in the Yucca Mountain TSPA-VA (after US Department of Energy, 1998). Comparison of base case expected value K_c with the base case 5th and 95th percentile values.*

4 Recent Observations of Radionuclide Transport Associated With Colloids at the Nevada Test Site

Recently, US DOE researchers at the NTS have demonstrated migration of Pu 1.3 km from the site of one of the underground nuclear tests, and have suggested that the migration may have occurred owing to the attachment of the Pu to colloids. These observations are summarised in this section; the relevance of these observations to the situation in Sweden is discussed in Section 5.

4.1 The Nevada Test Site

4.1.1 Overview

The NTS is located in the Southern Great Basin, encompassing about 3,600 km² of southwestern Nevada, and is about 150 km northwest of the city of Las Vegas. The geological structure is dominated by north-south trending faults, and shallowly dipping Tertiary ash-flow tuff and lava units deposited on Palaeozoic basement. The volcanic rocks include porous, non-welded ash-flows, ash-falls, reworked bedded tuffs, massive highly-brittle welded ash-flows, and ash-fall rocks. Many of the thicker units have densely welded interiors, although substantial portions of the original volcanic glass have been diagenetically altered to hydrous assemblages dominated by zeolites (e.g., heulandite).

The present climate at the NTS is sufficiently arid that most of the precipitation received at the surface is returned to the atmosphere by evaporation and only a small fraction penetrates through the partially-saturated zone. The water table is deep, typically located 400 to 800 m below the ground surface. The water table is also shallowly dipping, and the large variation in depth to the water table across the area reflects the local topographic variation.

4.1.2 Radionuclide Production and Environmental Contamination during an Underground Nuclear Test

A nuclear explosion in the 10-1000 kiloton range may produce several Curies of fission products. During an underground nuclear test, the temperature may locally exceed 10⁶ K and about 70 tonnes of rock are vaporised, and another 700 tonnes of rock are melted, for every kiloton of explosive yield. Within this mass is incorporated about 60 g of radioactive material per kiloton yield (Smith, 1995). Most of the refractory radionuclide species, including rare earth elements, alkaline earth elements and actinides (except uranium) are incorporated into the melt glass that coalesces at the bottom of the underground cavity formed by the explosion. More volatile elements, such as alkali metals, U, Sb, I, and Ru, and gases such as Ar, Kr, Xe, are more widely distributed in the

cavity and the rubble chimney that is created directly above the cavity and which extends to the surface.

The pressures generated by an underground nuclear explosion may create fractures, or open pre-existing fractures, in the surrounding rock. Fission products may be injected into these fractures by a process known as “prompt injection”. Prompt injection has been demonstrated to operate over distances at least as great as several tens of metres (Nimz and Thompson, 1992; Smith *et al.*, 1996). It is unclear whether prompt injection involves instantaneous injection of a plasma into fractures immediately after detonation, or by flow of gases and fluids along fractures at lower temperatures and over longer periods (seconds or minutes) after detonation. Late-stage gaseous transport has been invoked to explain strong $^{137}\text{Cs}/^{90}\text{Sr}$ fractionation in fracture deposits adjacent to the INGOT nuclear test (Smith *et al.*, 1996).

Fission products will tend to dissolve in groundwaters coming into contact with the melt glass or other condensed solids derived from an underground nuclear test. However, after several years, all short-lived radionuclides will have decayed significantly; there will typically be less than ten fission product nuclides remaining in detectable concentrations from a given nuclear test. Furthermore, many radionuclides sorb strongly to the local wall rock, which further reduces the long-term threat to groundwater. Consequently, only a small fraction of the fission products are typically able to migrate away from the test site, with tritium being the most mobile in groundwater.

Because most fission products have remained close to the source, the level of radioactivity observed in NTS groundwaters is variable, but typically very low, and measured in picoCuries (pCi) per gramme or per ml (10^{-12} Curies per gramme or per ml). The average background radioactivity of NTS groundwater comprises 0.01 pCi of dissolved ^{40}K and 0.01 pCi of tritium, although the tritium content may locally range up to 1 pCi. Water containing up to 20 pCi per gramme or mL is considered safe for drinking. For comparison, a gramme of NTS volcanic tuff typically contains about 20,000 ppm K, 4 ppm U and 20 ppm Th (Broxton *et al.*, 1986), corresponding to a natural radioactivity of 30 pCi of ^{40}K , and about 20 pCi each of U (plus daughters) and Th (plus daughters).

4.1.3 DOE Groundwater Monitoring

There were 828 nuclear devices detonated underground at the NTS in the period 1962 to 1992, most of them located in the Yucca Flat and Pahute Mesa regions (Figure 4.1). About 30% of the tests were at or below the water table, and therefore radionuclide fission products derived from these explosions have the potential to contaminate groundwater reservoirs. The scarcity of groundwater resources in southern Nevada, and the need to ensure that drinking water supplies for NTS operations are not jeopardised, has led the DOE to adopt a groundwater protection policy (Aquilina, 1991).

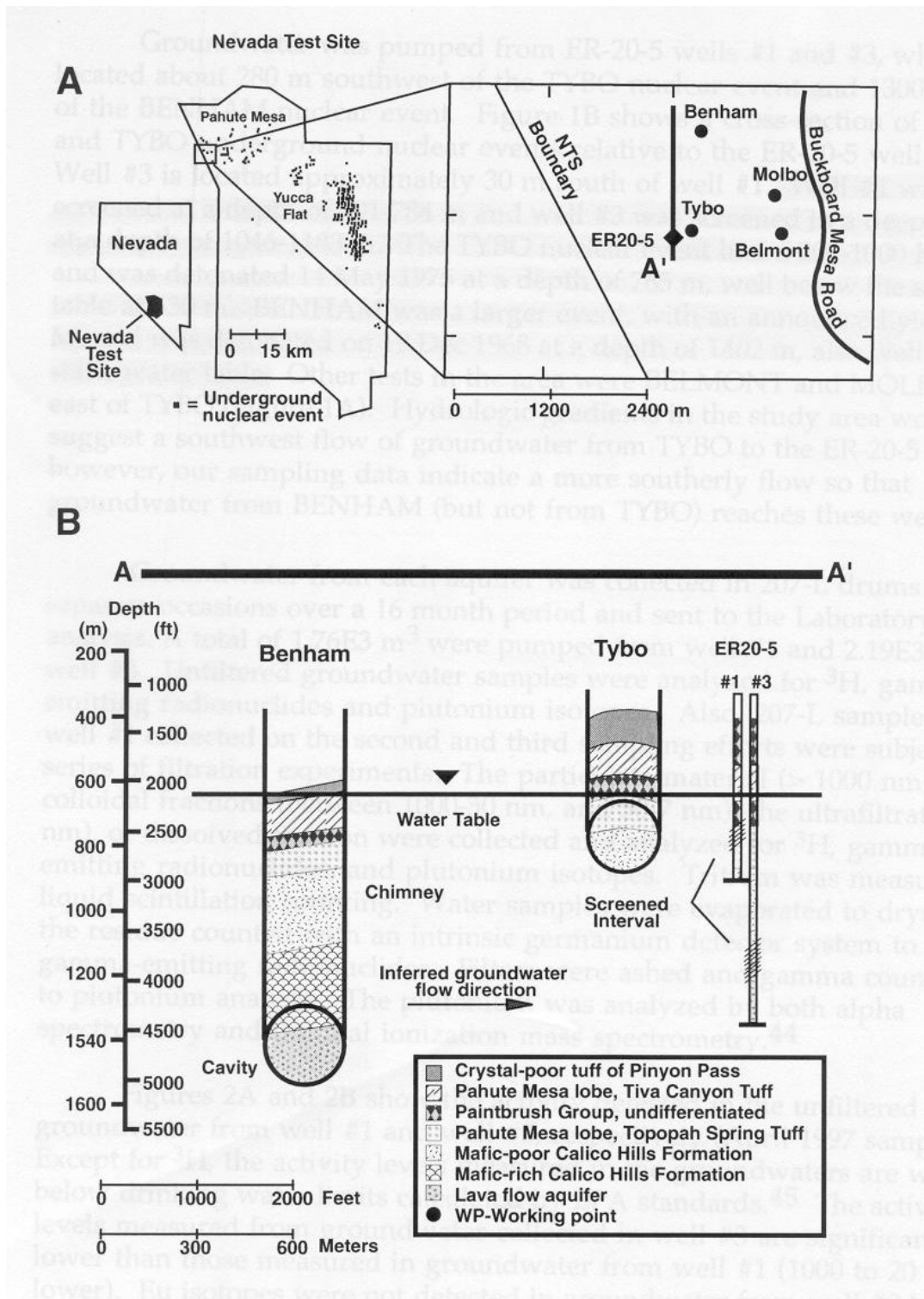


Figure 4.1: (A) Map of the NTS showing the location of underground tests in the Pahute Mesa region (after Thompson, 1998). (B) North-south geological cross section showing the volcanic stratigraphy at the BENHAM and TYBO test sites and the location of the ER20-5 well cluster (after Thompson, 1998).

Since 1973, the DOE Nevada Operations Office, Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) have been studying the effects of underground nuclear weapons testing on groundwater quality, and evaluating

the evidence for radionuclide migration in the subsurface. This programme now also involves the US Geological Survey (USGS), the Desert Research Institute (DRI) and contractors working at the NTS and is referred to as the Hydrologic Resources Management Programme (HRMP). The HRMP is sponsored by the DOE as part of the Environmental Restoration programme. The HRMP provides assistance in determining the current status of land and water resources at the NTS, and in predicting the future impact of radionuclide migration.

Several studies of radionuclide production and behaviour in the underground environment of the NTS, of the geological and hydrological consequences of nuclear testing, and of the factors controlling transport of radionuclides by groundwater have been carried out since 1973. A large number of test boreholes have been drilled for the purpose of sampling groundwater, and documenting the extent of radionuclide contamination and the source of the radionuclides (Nimz and Thompson, 1992). In the period November 1995 to February 1996, two holes were drilled adjacent to the site of the nuclear test TYBO. Water samples from these sites were subsequently found to contain radionuclides not often identified in previous studies. This finding prompted more detailed studies, described below, which provided evidence for km-scale migration of Pu in the natural environment.

4.2 Observations of Colloids and Radionuclides at the TYBO-BENHAM Site

4.2.1 Regional Setting of the TYBO-BENHAM Site

The TYBO nuclear test had an unclassified yield range of 200-1,000 kilotons and was detonated on 14 May 1975 at a depth of 765 m, well below the water table at 630 m. The BENHAM test was a larger event, with a reported yield of 1.15 Megatons, and was detonated on 19 December 1968 at a depth of 1,402 m, also well below the water table.

The TYBO-BENHAM site is located in the Pahute Mesa region, in the northwestern corner of the NTS, where thick sequences of ash-flow tuffs and rhyolitic lava flows dominate the geology. The volcanic rocks are fractured and faulted and form a series of distinct hydrologic units. Most of the faults follow the regional trend of Basin and Range faulting and strike north-northeast and north. Groundwater at Pahute Mesa migrates southwestward and southward. Estimated groundwater flow velocities are not well constrained and range between 1 and 80 m/yr (Blankennagel and Weir, 1973).

In November 1995, the Underground Test Area Operational Unit (UGTA OU) of the Environmental Restoration programme drilled well ER-20-5 #1 278 m southwest of the TYBO nuclear test and 1300 m south of the BENHAM nuclear test (Thompson, 1998; see Figure 4.1). ER-20-5 #1 was drilled to a depth of 860 m, cased and screened for water withdrawal in a transmissive aquifer at a depth of 701-784 m, a depth comparable to the working point (the point at which detonation occurred) of the TYBO nuclear event. In February 1996, well ER-20-5 #3 was drilled about 30 m south of ER-20-5 #1. ER-20-5 #3 was drilled to a depth of 1309 m, cased and screened for water withdrawal in

a transmissive aquifer at a depth of 1046-1183 m, a depth 200-300 m shallower than the working point of the BENHAM nuclear event.

4.2.2 Analyses of Groundwater, Soil and Melt Glasses

Groundwater was pumped from ER-20-5 #1 and ER-20-5 #3 and collected in 207-litre drums (Thompson, 1998). Collections were made on three separate occasions over a sixteen-month period. A total of 1,760 m³ was pumped from Well #1 and 2,190 m³ from Well #3. Unfiltered groundwater samples were analysed for tritium, gamma-emitting radionuclides, and Pu isotopes. Analyses of water from both wells recorded the presence of tritium, ¹³⁷Cs, ⁶⁰Co, ²³⁹Pu, and ²⁴⁰Pu, and at Well #1 only, the europium (Eu) isotopes ¹⁵²Eu, ¹⁵⁴Eu and ¹⁵⁵Eu (see Figure 4.2). Analyses also revealed that for a given well, the radionuclides present and their concentration were similar for each of the three sampling exercises, but that the concentrations of radionuclides in the shallower well #1 were several orders of magnitude higher than those measured in the deeper well #3. Except for tritium, the activity levels measured in the groundwaters were well below drinking water limits stipulated by the US Environment Protection Agency (EPA).

Whereas tritium, ¹³⁷Cs and ⁶⁰Co have often been observed in NTS groundwaters adjacent to underground nuclear tests, the presence of europium (Eu) and Pu in the groundwater samples was unexpected (Thompson, 1998; Kersting *et al.*, 1999). Eu isotopes are relatively insoluble and, where they had previously been observed at the NTS, they had been associated with colloids. Pu isotopes had never previously been encountered in NTS groundwaters remote from the working point of an underground nuclear test. The 207-litre samples collected in the second and third sampling efforts were subjected to a series of filtration experiments to check for the presence of colloids, and to determine whether the radionuclides were associated with the colloids. The following particulate size fractions were separated: >1000 nm, 1000 to 50 nm, 50 to 7 nm, and <7 nm (the ultrafiltrate or dissolved fraction).

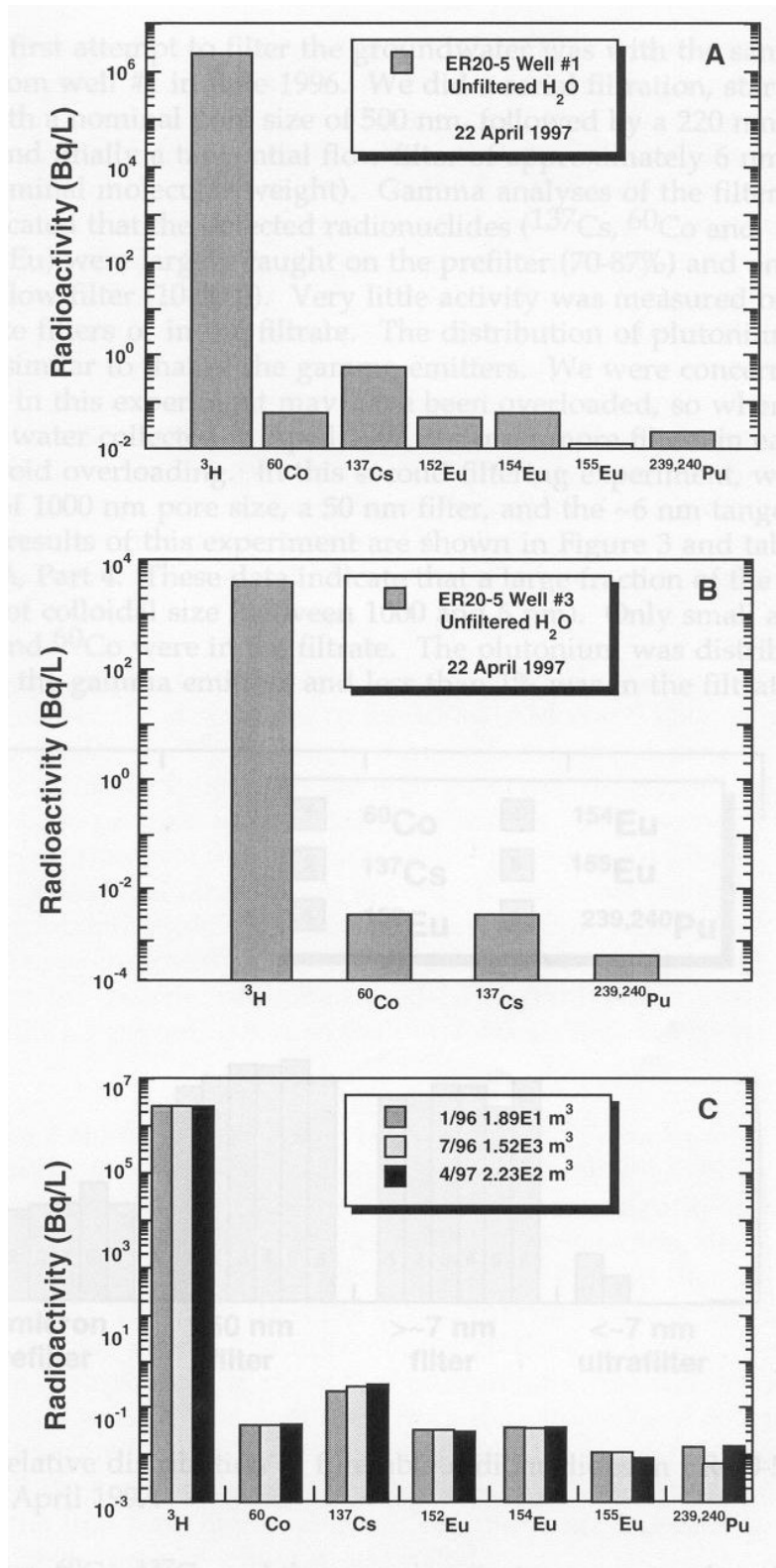


Figure 4.2: Isotopic data for groundwaters sampled at the ER20-5 well cluster (after Thompson, 1998).

The ultrafiltrates were analysed for tritium by liquid scintillation counting, and then evaporated to dryness and the residues analysed by gamma- and alpha-spectrometry (Thompson, 1998; Kersting *et al.*, 1999). The Pu was also analysed by thermal ionisation mass spectrometry (TIMS). Filters were analysed by gamma-spectrometry and alpha-spectrometry and by TIMS. The distribution of filterable radionuclides in ER-20-5 #1 water is shown in Figure 4.3, and indicates that most of the radionuclides were associated with the filtered material, and that this material is of colloidal size, between 1000 and 7 nm in size. Only small amounts of the ^{137}Cs and ^{60}Co and less than 1% of the Pu were observed to be in the filtrate.

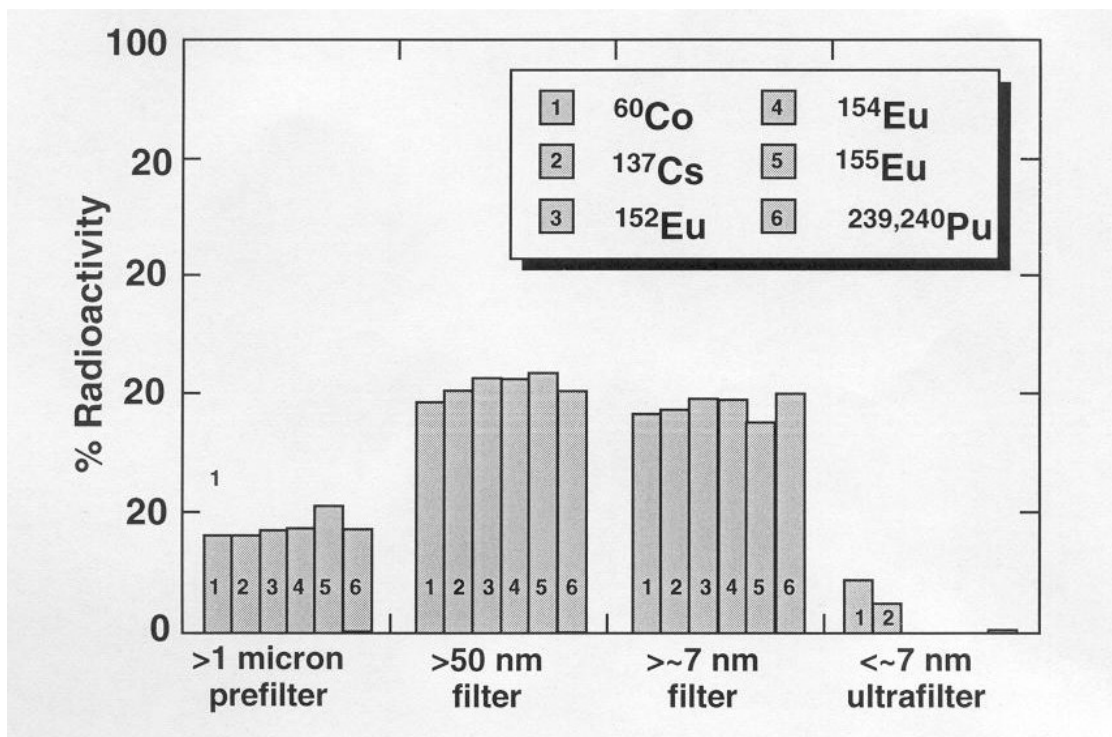


Figure 4.3: Distribution of filterable radionuclides in ER20-5 #1 well water collected in April 1997 (after Thompson, 1998).

In order to test for possible contamination of groundwater samples by airborne soil particulates, soil samples were collected from the vicinity of the ER-20-5 well cluster and analysed for Pu (Kersting *et al.*, 1999). The concentrations of Pu in soils was extremely low (<4 picogrammes Pu per gramme of soil), and the soil had a distinctly different isotopic composition from the groundwater samples (see Figure 4.4). Kersting *et al.* (1999) concluded that Pu detected in the groundwater samples did not derive from surface soil contamination.

4.3 Interpretation

4.3.1 Source of Radionuclides

Tritium, ^{137}Cs , ^{60}Co and the Eu isotopes are produced by all nuclear tests and their isotopic signatures cannot be used to determine their source. In contrast, the $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratio is unique to a specific nuclear event and, as a result, the Pu isotopic composition of the groundwater and filtered samples can be used to identify the source of the Pu (Thompson, 1998). Archived melt glass material collected from the cavity region immediately after the detonation of the BENHAM and TYBO events was analysed for its $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic composition. Samples were split and aliquots analysed at LANL and LLNL to provide independent verification. The two laboratories used different chemical separation protocols, thereby eliminating the potential for laboratory bias. Procedural blanks were also analysed. These blanks had Pu concentrations significantly below the concentrations analysed in the groundwater samples, and did not contribute to the isotopic ratio measured. Pu analyses agree within 1.5% between the two laboratories (Kersting *et al.*, 1999).

Figure 4.4 shows a comparison of Pu isotopic composition measured in groundwater, filtered material, soil and melt glass material from various underground tests in the vicinity of the ER-20-5 well cluster (Thompson, 1998). $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratios are classified information. The data are therefore reported relative to the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of unfiltered groundwater in Well #1. The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio of the unfiltered groundwater in Well #1 matches that measured in groundwater from Well #3, that measured on the colloidal fraction from Well #1, and that of the BENHAM event. The ratio does not match that of the TYBO event, or any other nuclear test up-gradient from the ER-20-5 well cluster, or that of the soil samples. The data demonstrate that the Pu at Well #1 and Well #3 is from the same source, and that the source is the BENHAM test site, 1.3 km to the north. No Pu from the TYBO nuclear test site is detectable in the sampled groundwater. Figure 4.4 demonstrates that Pu from any of the other possible sources is isotopically distinct and mixing between Pu from different sources would cause the groundwater samples to plot above the line in Figure 4.4.

The other radionuclides detected in groundwaters from the ER-20-5 well cluster cannot be shown to originate from any particular nuclear test. Thompson (1998) assumed that Eu, which is often used as a chemical analogue for actinides, and possibly the other radionuclides, also originated from the BENHAM test site.

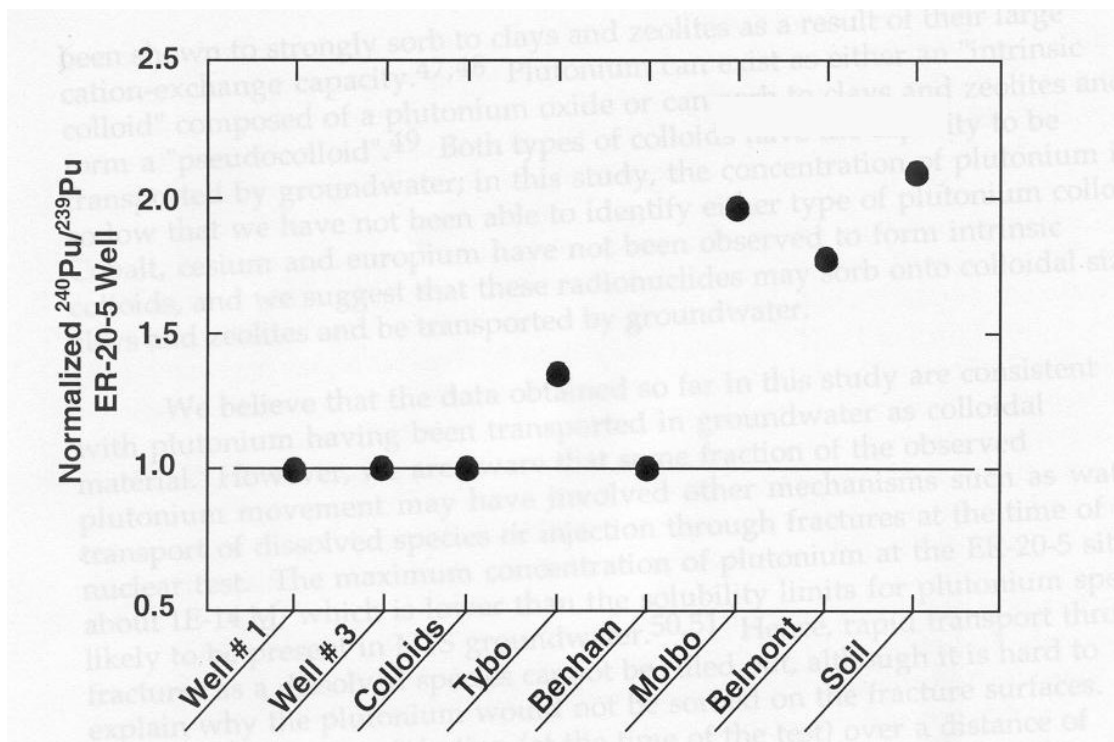


Figure 4.4: Isotopic ratios for $^{240}\text{Pu}/^{239}\text{Pu}$ in various samples from Pahute Mesa. All ratios are relative to that from the BENHAM event (after Kersting *et al.*, 1998). The uncertainty in each $^{240}\text{Pu}/^{239}\text{Pu}$ measurement is approximately ± 0.05 normalised $^{240}\text{Pu}/^{239}\text{Pu}$ units.

4.3.2 Association of Radionuclides with Colloids

The data presented in Figure 4.3 demonstrate the association of Co, Cs, Eu and Pu with colloid-sized particulates in the groundwater sampled from ER-20-5, Well #1 and Well #3. The different size fractions collected on the filters were analysed by x-ray diffraction (XRD) and with a scanning electron microscope (SEM). The colloidal material was composed of clay (illite), zeolites (mordenite and heulandite), cristobalite and minor glass. The same minerals were detected in each of the three size fractions: >1000 nm, 1000 to 50 nm, and 50 to 7 nm. Quantitative chemical analyses could not be obtained by SEM or electron microprobe. However, the observed mineralogy is consistent with the host-rock lithology (Kersting *et al.*, 1999).

Clays and zeolites are common secondary minerals in altered rhyolitic tuff and have been identified in the rocks of Pahute Mesa (US Department of Energy, 1998). The mineralogy and concentrations of colloids were determined in groundwaters collected in deep volcanic tuff aquifers located in the southwestern part of the NTS, away from the location of nuclear tests (Kingston and Whitbeck, 1991; Viani and Martin, 1996). These studies identified naturally occurring colloids composed of minerals similar to those

found by Kersting *et al.* (1999), namely clays (sheet silicates), zeolites (clinoptilites), and cristobalite.

Clays and zeolites have high sorption capacity for actinides and other cationic radionuclides. In some cases, the capacity of radionuclides to sorb to specific minerals is well known. For example, ^{137}Cs , ^{60}Co and actinides have been shown to sorb strongly to clays and zeolites as a result of their large cation exchange capacity. Pu can exist as either an “intrinsic colloid” composed of polymeric Pu oxide, or can sorb to clays and zeolites to form a “pseudocolloid”. Both types of colloid may be transported by groundwater. However, researchers at LLNL and LANL have so far been unable to identify either type of Pu colloid in the NTS groundwater sampled from ER-20-5. Co, Cs and Eu have not been observed in the form of intrinsic colloids in NTS groundwaters, and Thompson (1998) suggested that these radionuclides may sorb onto colloidal-size clays and zeolites and be transported by groundwater.

4.3.3 Possible Transport Mechanisms

The data reported above demonstrate that the Pu observed in the ER-20-5 wells was derived from the BENHAM test site 1.3 km to the north, but the data do not reveal how the Pu was transported. The Pu is now associated with the groundwater colloidal fraction, but this does not mean that it was transported for any or all of the entire 1.3 km as colloidal material.

Three Pu transport mechanisms are possible:

- Prompt injection of fission product material, either as a plasma, or in gaseous or liquid form, along fractures. This mechanism has been inferred to cause movement of fission products on a scale of tens or hundreds of metres (Nimz and Thompson, 1992; Smith *et al.*, 1996). However, because groundwater may also preferentially move through the same fractures, it can be difficult to determine conclusively whether the mechanism for radionuclide migration was groundwater movement or fracture injection.
- Transport of dissolved Pu species in groundwater. The maximum concentration of Pu at the ER-20-5 site is lower than the solubility limits for Pu species likely to be present in NTS groundwater. Hence, rapid transport through fractures as a dissolved species is possible. However, it is likely that most Pu would be sorbed onto fracture surfaces (Thompson, 1998).
- Transport of Pu associated with colloidal material. Co, Cs, Eu and Pu were associated with colloid-sized particulates in the groundwater sampled from ER-20-5. Colloidal radionuclide transport can occur either at faster or slower rates than the transport of dissolved radionuclides. Enhanced transport rates can result if colloids are mobile in parts of the hydrogeologic system where flow rates are greater than average. Fast flowing regions may occur in the centre of fractures or channels, for example. Colloids may tend to stay in such fast flowing regions if they are too big to enter the pores of the rock where flow is slower. Electrostatic repulsion between

colloids and the rock may also tend to keep the colloids in the fast flowing regions. The maximum effect of colloids in transporting radionuclides is likely to be observed if the colloids themselves are composed of radionuclides (e.g., actinide intrinsic colloids) or if the radionuclides are irreversibly bound to the surfaces of the colloids. Colloidal radionuclide transport rates may be reduced below their theoretical maximum if the colloids are destabilised under the prevailing geochemical conditions, if the colloids are physically filtered as they move through the rock, if the colloids interact chemically with, and bind to, the surface of the rock, or if the attachment of radionuclides to mobile colloids is not irreversible. However, at the NTS, the identity of the Pu-colloidal material, the distance over which Pu-colloidal material was transported and the details of how the Pu was transported, remain uncertain.

Thompson (1998) and Kersting *et al.* (1999) concluded that it was most likely that the Pu transport from the BENHAM site to the ER-20-5 well cluster had been facilitated by colloids. They dismissed transport as a dissolved species as unlikely, owing to the tendency of Pu to sorb to fracture surfaces. They argued that it was unprecedented to observe prompt injection over distances of 1.3 km, and moreover, the Pu would have to have been deposited in two separate aquifers separated by 300 m vertically and 30 m horizontally (see Figure 4.1).

The hydrology of the TYBO-BENHAM area is not well known and the most likely route taken by the Pu, if it was transported exclusively by groundwaters, is unclear. The activity measured in groundwater from the shallower aquifer (Well #1) is significantly greater than that measured in the deeper aquifer (Well #3), despite the shallower aquifer lying 600 m stratigraphically above the cavity of the Benham event, where most of the melt-glass resides. It is possible that some melt-glass was extruded up the chimney overlying the cavity to provide a source of Pu at higher structural levels. Alternatively, groundwater previously flowing at the BENHAM cavity depth may have been re-routed, moving upwards to more permeable zones via the rubble chimney. Such a phenomenon has previously been noted at the NTS at the CHESHIRE nuclear test site (Buddemeier, 1988).

We would note, however, that the regional trend of Basin and Range faulting is north-south, the same as the direction of migration from BENHAM to the ER-20-5 well cluster. Prompt injection may have been able to exploit north-south fault lines and facilitate emplacement of fission products towards the south. Furthermore, the BENHAM nuclear test was a large, deep detonation (1.2 Megatons) that would have generated a large volume of melt (~100 m diameter sphere). The BENHAM working point was below the water table and widespread vaporisation of pore fluids would have occurred. Together, these processes may have caused exceptional prompt injection transport distances.

Finally, we would also note that severe perturbations to the regional hydrologic system may be caused by underground testing. Repeated testing on Pahute Mesa after the BENHAM event could have had a significant impact on groundwater flow in this region.

In summary, the three potential transport mechanisms are not mutually exclusive and it is possible that the observed Pu transport involved a combination of the three mechanisms.

4.3.4 Remaining Uncertainties and Future Work

Many uncertainties remain in understanding of radionuclide migration and colloid transport at the NTS. Some of the most significant uncertainties are as follows:

- The groundwater transport pathway and distance that the Pu migrated are unknown.
- The maximum extent of Pu migration is unknown. Insufficient boreholes have been drilled to determine the current spatial distribution of Pu from the BENHAM test.
- It is not known if migration of Pu is unique to the BENHAM test.
- The hydrologic system in the TYBO-BENHAM area is not well known. In particular, it is not known why waters in the shallower aquifer at ER-20-5 have a higher Pu concentration, even though the working depth of BENHAM is closer to the depth of the deeper aquifer. It is also not known why material from the TYBO event is absent from the ER-20-5 well cluster, even though it is much closer to the well cluster than the BENHAM event.
- It is not known how the radionuclides are associated with colloids, or whether the Pu exists as intrinsic colloids or pseudocolloids.

In addition to these uncertainties, there are also unresolved questions concerning colloid concentrations, size distributions, flow in fractured media, and exchange of radionuclides between colloid and fracture surfaces, all of which may influence the ability of colloids to transport radionuclides at the NTS.

4.4 Summary

Groundwaters sampled at the NTS ERDA 12 well cluster contain radioactive isotopes of Pu, Am, Cs, Co and Eu. The isotopic composition of the Pu observed at the ERDA 12 well cluster matches the composition of Pu from the BENHAM underground nuclear test, implying that the Pu has migrated 1.2 km in ~30 years.

At the sampling point, the radionuclides are associated with colloidal material in the groundwater. However, the identity of the colloidal material is uncertain. Cs, Co and Eu are probably associated with clays and zeolites, whereas Pu may be an intrinsic colloid. Ongoing US DOE research is seeking to determine the nature of the colloidal material.

It is not clear whether the Pu has been transported as colloids for the entire distance from the BENHAM working point to the observation point. The mechanism of prompt

injection has been observed to transport radioactive material outward from an underground nuclear test shot point at other NTS localities, but has never before been observed to operate on a scale greater than 300 metres. The high permeability of some volcanic units such as lava flows, and the perturbations to the regional hydrologic system caused by repeated underground nuclear testing, may be responsible for enhanced radionuclide transport.

The NTS observations suggest that Pu transport models taking into account only sorption and solubility may underestimate Pu migration potential. As a result of these observations, PAs for the Yucca Mountain Site have taken into account the potential for colloidal transport of actinides, and have used wide ranges of parameter values to assess the remaining uncertainty. The TSPA-VA has included mechanistic models, and dose calculations for the transport of radionuclides by colloids involving both reversible and irreversible sorption.

5 Comparison of NTS Observations and KBS-3 Concept

In this Section, we evaluate the relevance of the NTS observations described in Section 4 to the KBS-3 disposal concept. In particular, we compare the NTS geo-hydrological environment to the KBS-3 environment.

5.1 KBS-3 Disposal Concept

The KBS-3 repository for spent nuclear fuel is composed of a system of barriers:

- The fuel is placed in copper canisters. Inside the five-m-long canisters is a cast iron insert to provide mechanical strength.
- The canisters are surrounded by a layer of bentonite clay. The clay provides mechanical protection, and also limits access of groundwater and corrosive substances to the canisters. The bentonite also adsorbs radionuclides that may be released from the canisters, and filters colloids that may form within the repository.
- The canisters and surrounding bentonite clay are emplaced at a depth of about 500 m in crystalline bedrock, where mechanical and chemical conditions are stable.

5.2 NTS/KBS-3 Comparison

There are several comparisons that can be made between the KBS-3 repository environment, and the environment in which the NTS colloid observations were made. These include:

- **Geological comparison:** The KBS-3 geological host environment consists of fractured granite whereas at Yucca Mountain it consists of volcanic ash-flow tuffs having various degrees of welding.
- **Hydrological comparison:** The KBS-3 environment is entirely below the water table and any transport of radionuclides is assumed to occur under water-saturated conditions. At the NTS, both the source of radionuclides and the sampling point are also below the water table.
- **Colloid comparison:** For typical deep groundwaters in Swedish fractured granite, the colloids are composed of iron hydroxides, clay minerals and silica minerals. At the NTS, colloid types identified include the same three phases and also zeolites. The breakdown of volcanic glass in NTS rocks may provide a source of colloids that does not exist in the KBS-3 environment. However, the concentration of colloids in NTS and Swedish groundwaters is similar (see below).

- **Emplacement comparison:** The emplacement of the radionuclide source material is passive for the KBS-3 concept, whereas for NTS radionuclide material it is explosive. The source isolation for KBS-3 involves an extensive engineered barrier system. At the NTS, there is no barrier system for radionuclide source material.

A more detailed comparison is provided below.

5.2.1 NTS/KBS-3 Geological Comparison

The mineralogy of the host rock at both sites is similar, although there are major textural differences. Although the KBS-3 geological environment consists of crystalline granitic bedrock, as compared with variably welded silicic ashflow tuffs and lava flows at the NTS, both rock-types are dominated by a quartzofeldspathic mineralogy. In addition, NTS rocks contain variable amounts of volcanic glass, and low temperature breakdown products of this glass such as clay minerals and zeolites.

The main textural difference between KBS-3 and NTS is that the NTS volcanic rocks are finer grained and more heterogeneous. Locally, the NTS rocks contain abundant volcanic glass where ash flow tuffs have been strongly welded. The higher the degree of welding, the more consolidated and massive the rock, and the lower the permeability. The NTS rocks generally comprise layers of both strongly welded, low-permeability units such as the Topopah Spring Tuff, and essentially unwelded units such as rhyolitic lava flows. The KBS-3 host rock consists of coarsely crystalline granitoids which, away from fault zones, are relatively homogeneous and texturally isotropic. The KBS-3 geological environment is therefore more homogeneous than the NTS environment, in terms of both texture and mineralogy.

5.2.2 NTS/KBS-3 Hydrological Comparison

Fracture flow dominates within the saturated zone both at the NTS and for the KBS-3 concept. However, as explained in the previous section, the NTS rocks are texturally more heterogeneous, and this also applies to their hydrological properties. For KBS-3, permeability in the host rock is assumed to be uniformly low (SKB, 1999a), whereas at the NTS, there is considerable variation in permeability, with some highly transmissive units such as lava flows interspersed with highly impermeable densely welded units.

In terms of hydrological regime, KBS-3 is assumed to be steady-state, whereas at the NTS, transient effects are caused by underground detonations. It is known that such detonations can significantly perturb hydraulic heads for periods of up to several years in the region close to a large underground test detonated below the water table. This phenomenon may be significant in the case of the observations in the TYBO-BENHAM region. The TYBO event post-dated the BENHAM event, which is the source of the Pu sampled in groundwater. It is possible that the TYBO detonation influenced the groundwater flow in the region and facilitated transport of colloidal material and/or Pu from BENHAM to the sampling point.

5.2.3 NTS/KBS-3 Colloid Comparison

The presence of volcanic glass and its devitrification products, including minerals such as zeolites and clay minerals that often tend to form colloids, suggests that the NTS rocks may be a more likely source for colloids than the KBS-3 granitic host rock. However, measurements of the concentration of colloids in deep Swedish groundwaters, and at the NTS and Yucca Mountain, are similar:

- Laaksoharju *et al.* (1995) reported inorganic colloid concentrations of 20 to 45 µg per litre (ppb) in saline and non-saline Swedish groundwaters, sampled from between 70 and 1600 m below the surface. These concentrations apply to colloids in the size range 50 to 400 nm.
- Triay *et al.* (1996) measured colloid concentrations in Yucca Mountain groundwater sampled from 500 to 1000 m below the surface, and obtained values in the range 10 to 30 µg per litre. These concentrations apply to colloids in the size range 100 nm to 10µm.

Therefore, there appears to be no major difference in the natural background colloid concentration at the two sites.

Colloid types observed in deep Swedish groundwaters, and at the NTS and Yucca Mountain, are also similar. In Sweden, the main colloid types observed include silica, clay minerals and iron oxyhydroxides (Laaksoharju *et al.*, 1995), and similar colloid types, and also colloidal zeolites, were observed in Yucca Mountain groundwaters (Triay *et al.*, 1996; US Department of Energy, 1998).

5.2.4 NTS/KBS-3 Emplacement Comparison

It is hard to imagine a greater contrast than between a nuclear explosion, which disperses material with great force, and the KBS-3 disposal concept, which is designed to isolate the waste for long periods. At the NTS, the radionuclide source was actively dispersed by a nuclear explosion that generated fractures, which radiated from the working point, and a collapse chimney, which extended vertically to the surface. For KBS-3, it is assumed that the radionuclides are emplaced within copper canisters, which are then sealed within the repository using bentonite and cementitious materials. Pu-colloid migration could only become important after the engineered barrier system was breached or significantly degraded.

5.3 Summary - Are the NTS Observations Relevant to Sweden?

At the NTS, colloids may have promoted ≥ 1.3 km migration of Pu and other radionuclides in 30 years. The following comparison between the KBS-3 disposal concept and the NTS environment has been made:

- The mineralogy of the KBS-3 geological environment and the NTS is similar. However, NTS rocks are finer grained and more heterogeneous in composition and texture.
- The KBS-3 and NTS hydrological environments are dominated by fracture flow. KBS-3 is assumed to be in a steady-state hydrological regime, whereas at the NTS, transient effects are caused by underground detonations.
- There is no major difference in the natural background colloid concentration, or the main colloid types at the NTS or in the KBS-3 concept.
- A key difference between the NTS and KBS-3 is the contrasting mode of emplacement of the radioactive material. In the KBS-3 disposal concept, the waste is emplaced passively, and is protected from the surrounding geo-hydrological environment by an engineered barrier system. At the NTS, the radioactive source material was emplaced explosively, with attendant fracturing of the local country rock, and it was immediately exposed to flowing groundwater. In the KBS-3 disposal concept, the waste would only ever become directly exposed to groundwater after the engineered barriers were breached.

6 Evaluation of SKB's Treatment of Colloids in SR-97 and SFL 3-5

In this section we provide an evaluation of SKB's treatment of colloids in its two recent PA studies, SR-97 (SKB, 1999a; SKB, 1999b) and SFL 3-5 (SKB, 1999c). We discuss briefly SKB's assumptions about colloids and identify activities that may need to be conducted to support the waste disposal concepts further.

We first consider colloid issues that are common to both SR-97 and SFL 3-5 and, in particular, the occurrence and stability of natural groundwater colloids in the geosphere (Section 6.1). We then discuss issues relating to the individual performance assessments and disposal concepts - for example, waste and repository-derived colloids (Sections 6.2 and 6.3).

6.1 Groundwater Colloids

In both SR-97 and SFL 3-5, SKB assumed that groundwaters at the repository site will be saline. SKB assumed that the salinity of the groundwaters will destabilise colloids by the process of coagulation, so that only a low concentration of colloids will remain dispersed or suspended in the groundwater. SKB argued, therefore, that groundwaters entering the repository will introduce only a small concentration of colloids. SKB has evidence that typical Swedish groundwaters are saline and contain low concentrations of colloids (Laaksoharju *et al.*, 1985). To ensure that groundwater colloid concentrations are sufficiently low, Laaksoharju *et al.* (1985) recommended that groundwaters at the repository site should contain at least 10^{-4} mol/l of dissolved calcium (Ca^{2+}).

We would note, however, that 10^{-4} mol/l is lower than the ionic strength of Yucca Mountain groundwaters under ambient conditions. For typical Yucca Mountain groundwaters having a composition of 5×10^{-3} molal (CRWMS M&O, 1998), a colloid concentration of about 6×10^{-5} mg/ml, or 7×10^6 particles per ml, was estimated. These parameters were used as input to the TSPA-VA colloid sorption and transport model for Pu.

To support its assumptions regarding groundwater colloids, SKB will need to demonstrate that groundwater compositions at the site selected for the repository are indeed such that the concentrations of groundwater colloids are sufficiently low. SKB will therefore need to conduct a repository site characterisation programme that includes adequate characterisation of groundwater compositions and colloid contents.

We also note the possibility of scenarios in which colloids may become relatively more significant. For example, glaciation may lead to the influx of relatively oxidising, fresh groundwaters; such changes in groundwater geochemistry may increase colloid concentrations, stability and mobility. These effects may warrant further examination when assessing the performance of the proposed disposal system.

6.2 Colloids in SR-97

6.2.1 Scenarios and Colloidal Radionuclide Transport in SR-97

SKB considered five scenarios in SR-97:

Base Scenario	Repository built as designed; constant climate; no canister defects.
Canister Defect Scenario	Repository built as designed; constant climate; a few canisters with initial defects.
Climate Scenario	Repository built as designed; climate change leading to glaciation. Variants of this scenario are considered with and without canister defects.
Earthquake Scenario	To explore the potential for canister damage as a result of earthquakes.
Intrusion Scenario	To illustrate the potential consequences of future human activities.

Although no criteria are specified for the bentonite buffer relating to colloid filtration (SKB, 1999a), SKB places reliance on the bentonite to act as an effective filter preventing the escape of colloidal radionuclides from the near-field to the geosphere (SKB, 1999b). Recent experiments support the assumption that the bentonite buffer will act as an efficient colloid filter (e.g., Kurosawa *et al.*, 1997) and consensus appears to exist within the radioactive waste disposal community that this assumption is reasonable (Hadermann *et al.*, 1998). Thus, for scenarios in which the buffer remains intact, the approach taken by SKB to eliminate the effects of waste- and repository-derived colloids from the PA calculations is reasonable. There may, however, be a case for examining in more detail the potential role of groundwater colloids in enhancing radionuclide transport, particularly in the climate change scenario in which glaciation may affect colloid concentrations, stability and mobility.

For scenarios in which the buffer may be degraded, there is also a potential need to account for colloids in the PA. Two of the SR-97 scenarios involved failure of the buffer: the earthquake scenario and the human intrusion scenario. Other causes for buffer failure such as gas production can be envisaged, and may in future require more detailed treatment.

Results from the SR-97 analyses indicate that there is a low probability that a small number of canisters might be damaged as a result of the effects of an earthquake. SKB provided little or no discussion of the potential effects of an earthquake on the bentonite buffer. SKB recognised that the approach used in SR-97 for assessing the earthquake scenario is subject to considerable uncertainties (SKB, 1999a). In light of the

uncertainties associated with the SR-97 analysis of the potential effects of earthquakes, radionuclide transport calculations were not performed for the earthquake scenario. SKB is refining its approach to assessing the potential effect of earthquakes and suggests that a refined approach may be used in future PAs. Should SKB's refined approach indicate that an earthquake could lead to the release of radionuclides from a damaged canister, and that the buffer's capacity to filter colloids might be reduced, even temporarily, as a result of structural deformation prior to the buffer re-sealing, then it may be appropriate to undertake radionuclide transport calculations including colloids.

As part of the SR-97 analyses, SKB has undertaken calculations to illustrate the potential consequences of inadvertent penetration of a waste canister by a borehole. SKB considers two potentially exposed groups: drilling personnel and a family that settles at the site after the drilling event. The effects of radionuclide transport by waste- and repository-derived colloids on doses to drilling personnel will be negligible when compared to the effects of radionuclide transport by exhumation of solid waste material. The basis for eliminating waste- and repository-derived colloids from radionuclide transport calculations made to evaluate the potential doses to a family that settles by an abandoned borehole is not clear. The SR-97 analysis is based on several pessimistic assumptions, including that radionuclides reaching the biosphere are concentrated in peat and are not subsequently dispersed by, for example, humic-colloid transport. However, there may be a case for including waste- and repository-derived colloids in calculations of the amount of radionuclides that may be transported to the biosphere via the borehole.

6.3 Colloids in SFL 3-5

SKB's preliminary safety assessment for the future low-level and intermediate-level waste repository, SFL 3-5 (SKB, 1999c), makes use of SR-97 (SKB, 1999a) and data from the Aberg, Beberg and Ceberg sites.

The potential effects of colloids on radionuclide transport from the low-level and intermediate-level waste repository were not evaluated quantitatively in the SFL 3-5 preliminary safety assessment. SKB argued that the effects of colloids can be excluded from PA calculations because the concentrations of colloids in the waters of the disposal system will be low. SKB also suggested that colloid and colloid-facilitated radionuclide transport may be limited.

6.3.1 Colloid Formation in SFL 3-5

Cement-Derived Colloids

SKB assumed that only low concentrations of colloids will be produced from the cementitious materials in the SFL 3-5 repository. SKB based this assumption on studies of the Maqarin natural analogue site, where only low colloid concentrations have been observed in high-pH groundwaters that are similar to those expected in the SFL 3-5 repository.

To support its assumptions regarding the formation of colloids from cementitious materials, SKB will need to demonstrate that the potential for colloid generation from cementitious repository materials is indeed low. This may necessitate SKB conducting an appropriate experimental programme using repository-specific cementitious materials.

Other Colloid Sources and Types

SKB did not investigate all of the possible colloid formation processes that may occur in and around the SFL 3-5 repository. SKB's position is appropriate for a preliminary safety assessment. For a complete safety case, however, SKB would be expected to identify and discuss all of the potentially significant sources and types of colloids. Colloids may be formed by several processes, including waste degradation and chemical precipitation at steep chemical gradients within the repository system (e.g., at the boundary between the high-pH near-field and the near-neutral far-field). Colloid types include actinide intrinsic colloids (e.g., polymeric plutonium), inorganic colloids (e.g., mineral fragments), organic colloids (e.g., humic and fulvic acids), and microbes (e.g., bacteria).

SKB argued that the ultimate control on the significance of colloids is groundwater salinity and colloid stability (see Section 6.1). However, SKB will need to conduct an appropriate research programme into colloid formation and stability that considers a range of formation processes and repository-specific materials, including the wastes and waste container materials. SKB may also need to conduct research into colloid generation associated with chemical gradients within the disposal system.

6.3.2 Colloidal Radionuclide Transport in SFL 3-5

Anion Exclusion

SKB argued that the transport of colloids from the waste may be limited by anion exclusion. The surfaces of the colloids and pores of the waste container and backfill materials are likely to be negatively charged. SKB (Wiborg, 1995) argued that electrostatic repulsion between the colloids and the pores will limit colloid transport. Wiborg (1995) referred to results presented by Torstenfelt (1986) on the migration of fission products and actinides in compacted bentonite and, in particular, on the low diffusivity observed for iodide (I⁻).

SKB's argument (SKB, 1999c) that the transport of colloids from the waste may be limited by anion exclusion is questionable. The repository design described in the preliminary safety assessment for the SFL 3-5 repository includes a backfill of crushed rock. In contrast, Wiborg (1995) considered a repository in which the backfill was compacted bentonite. Since crushed rock has properties that are significantly different to those of compacted bentonite, the relevance of Torstenfelt's (1986) observations on the diffusivity of iodide to the SFL 3-5 design is not clear.

Competitive Sorption

As described above, SKB expects only low concentrations of colloids in the SFL 3-5 disposal system (SKB, 1999c). Based on work performed in relation to the disposal of spent fuel (Allard *et al.* 1991), SKB argued that the capacity of colloids in the SFL 3-5 disposal system to transport radionuclides will be small in comparison with the ability of the host-rock to retard radionuclide transport by sorption. Further support for this argument could be derived by applying the type of analysis undertaken by Allard *et al.* (1991), for spent fuel, to the specific characteristics of the SFL 3-5 repository and relevant waste types.

Gas Effects

With reference to the disposal of spent fuel, Neretnieks and Ernstsson (1997) proposed a link between gas bubble migration and colloid transport. SKB did not discuss the interaction between colloid transport and gas migration (SKB, 1999c). The potential for gas generation in the SFL 3-5 repository for low-level and intermediate-level wastes is likely to be greater than that in a repository for spent fuel. SKB ought, therefore, to evaluate the potential significance to the disposal of low-level and intermediate-level waste of the interaction between colloid transport and gas migration.

6.4 Summary

The KBS-3 concept includes a bentonite buffer that is likely to prevent the release of waste- and repository-derived colloids from the repository. An additional barrier to colloidal radionuclide transport may be provided if the repository is sited at a location where groundwaters in the host rock remain sufficiently saline that they destabilise colloidal species. However, there may be justification for performing further work to examine in more detail the potential role of groundwater colloids in enhancing radionuclide transport, during a scenario in which glaciation occurs. It would also be reasonable to include the effects of waste- and repository-derived colloids in calculations of the amount of radionuclides that may be transported to the biosphere following an earthquake that damages the engineered barriers, or the abandonment of a borehole that intersects the waste.

The potential significance of colloidal radionuclide transport to the SFL 3-5 concept for the disposal of low-level and intermediate-level waste is uncertain. In contrast to the KBS-3 concept, the SFL 3-5 repository does not incorporate a bentonite buffer and, therefore, there is a greater chance that waste- and repository-derived colloids may contribute significantly to radionuclide transport. In addition, when compared to the KBS-3 concept, the SFL 3-5 repository will contain a greater range of waste materials from which colloids may be derived by a range of degradation processes. Furthermore, some of these degradation processes (e.g., gas generation) may occur to a greater extent in the SFL 3-5 repository than in the KBS-3 repository. Further work appears to be justified to:

- Study the potential effects of colloid generation in the SFL 3-5 repository considering the range of potential colloids formation processes and repository-specific materials, including the wastes and waste container materials.
- Study the potential effects of colloid generation associated with chemical gradients within the SFL 3-5 disposal system.
- Apply the type of analysis undertaken by Allard *et al.* (1991), for spent fuel, to the specific characteristics of the SFL 3-5 repository and relevant waste types.
- Evaluate the potential significance to the disposal of low-level and intermediate-level waste of the interaction between colloid transport and gas migration.

7 Implications and Recommendations

In this Section, we consider the implications of the recent PAs and colloid observations for radioactive waste disposal in Sweden, and make recommendations regarding the need for further assessment of colloids in the KBS-3 concept.

7.1 Treatment of Colloids in Swedish PA: Historical Summary

SKB has considered the potential importance of colloids in radionuclide transport in the context of both the KBS-3 and the SFL 3-5 disposal concepts, and has conducted supporting research (Allard *et al.*, 1991; Laaksoharju *et al.*, 1995). Although SKB has not evaluated colloidal transport of radionuclides directly in PA calculations, SKB has evaluated the impact of both reversible and irreversible sorption of radionuclides to colloids in supporting studies:

- In the case of reversible sorption, the impact of considering colloids led to a 1% change in the K_d values used. Such an effect would have a negligible impact on PA results.
- In the case of irreversible sorption, the combined maximum dose from the colloid fraction for all radionuclides was less than 1 mSv per year, even for a highly conservative scenario in which all colloids released from the repository are transported directly to a drinking water well (Allard *et al.*, 1991).

Consequently, calculations of radionuclide transport by colloids were not specifically included in recent SKB PAs (SR-95; SR-97; SFL 3-5). However, SKB maintains an experimental programme to address colloid transport issues, and has taken an active interest in US research connected with the NTS findings. A future experimental collaboration between SKB and LLNL researchers is planned, involving experiments to investigate colloid generation from bentonite at the Aspö underground research laboratory.

Colloid calculations were not specifically included in SKI's SITE-94 PA (SKI, 1996). However, SKI has developed a model for radionuclide transport by colloids, COLLAGE II (Grindrod and Cooper, 1993), and such a model could be developed further to evaluate the potential for Pu transport by colloids (see below).

7.2 Treatment of Colloids in Swedish PA: Future Approach

The following are possible methods of treating colloids in PA:

- (i) Development of qualitative screening arguments to eliminate colloidal processes from the PA based on literature review of existing information.
- (ii) Model development and application for the establishment of quantitative screening arguments to eliminate colloidal processes from the PA.
- (iii) Inclusion of colloidal processes in PA models.

The discussion in this section focuses on method (iii). All programmes that have modelled the transport of radionuclides attached to colloids have used a similar one-dimensional transport equation. In this respect, SKI's COLLAGE II model is an appropriate tool for use in PA calculations.

Most PAs that model colloid transport include irreversible sorption to colloids and instantaneously reversible sorption as modelling end-members (e.g. H-12, Nirex-97, TSPA-VA). Natural systems are likely to behave somewhere in between these end-members. In order to model sorption more realistically, models will need to consider the kinetics of binding. SKI's existing model of colloid transport, COLLAGE II, includes a capability for a kinetic approach. Therefore, a possible modelling strategy could be as follows:

Calibration of COLLAGE II

Calibrate the COLLAGE II model by making scoping calculations of colloidal Pu transport aimed at reproducing the NTS observations. As part of this calibration process the following parameters could be varied:

- Colloid concentration.
- Groundwater flow velocity.
- Porosity of rock.
- Fracture width and fracture density.
- Radionuclide-colloid partition coefficient.
- Radionuclide-host rock partition coefficient.
- Parameters constraining filtration or clogging of colloids.
- Rate of radionuclide sorption/desorption to colloids (kinetics of binding).

The objective would be to confirm that a suitably developed and calibrated COLLAGE II model is capable of reproducing the NTS observations, to constrain the possible parameter ranges required, and to compare the performance of COLLAGE II with the LANL calculations. This would build confidence in the use of COLLAGE II as part of SKI's independent PA capability.

Modelling evaluation of KBS-3 concept using COLLAGE II

Conduct structured sensitivity studies aimed at identifying potential conditions, for example critical modelling assumptions and areas of parameter space, in which colloids have the potential to influence the results of repository performance assessment calculations for the KBS-3 concept.

The COLLAGE II model could be used to model colloidal radionuclide transport for the KBS-3 repository concept taking into account the results from modelling the NTS observations, and using appropriate information on the characteristics of the potential repository sites considered in SR-97. To constrain the scope of the evaluation, a simplified representation of one of the SR-97 sites could be developed using the model and appropriate sensitivity studies made around that modelling base case.

The scenarios to be considered in the analyses would include a case in which the buffer remains intact and glaciation of the repository site causes an influx of relatively dilute waters to the groundwater system. Under these conditions the reduced salinity of the groundwaters may lead to increased colloid stability and mobility as compared to the SR-97 analyses. Although the evaluation would not necessarily extend to dose calculations, the sensitivity analyses would help to address the question of how much greater the groundwater colloid concentration would have to be to have a significant impact on disposal system performance. Ultimately, if the sensitivity analyses suggest that it is not possible for colloids to have a significant effect on calculated disposal system performance, it may be possible to eliminate such a scenario as a potential concern.

Comparison of colloid modelling approaches

An extensive modelling effort to evaluate colloid transport of radionuclides at the NTS and Yucca Mountain site is currently being carried out at LANL on behalf of the US DOE. A quantitative comparison between the assumptions and predictions of recently developed colloid transport models, with the assumptions and predictions of the new LANL work could be made. The objective would be to identify the extent to which the latest modelling work at LANL goes beyond or differs from work being carried out in radioactive waste disposal assessment programmes in countries outside the US.

The comparison could include the SKB colloid programme, results of the EC-sponsored CARESS project (Gardiner, 1997), the COLLAGE II scoping calculations, and the Yucca Mountain Viability Assessment, and could include:

- Comparison of colloid concentrations.

- Comparison of colloid types.
- Comparison of the assumed pseudo-colloid stability coefficients (K_d) for various key radionuclides.
- Assumptions and methods for representing fracture flow.
- Comparison of predicted colloid transport distances for reversible/irreversible models.

7.3 Summary Recommendations

The project findings lead us to make the following recommendations:

- Further work should be conducted to assess the potential significance of colloids to the performance of SKB's concepts for radioactive waste disposal, and SKB's work on colloids should be kept under review. The work should be directed at assessing the potential significance of colloids to the performance of both the KBS-3 disposal concept, and the SFL 3-5 disposal concept, in which there remain large uncertainties related to colloids.
- A "watching brief" should be maintained on future NTS developments. New results on NTS radionuclides and colloids will be available shortly from DOE's laboratory studies, field experiments, and modelling programme.
- Important lessons may be learned by monitoring developments in other PA programmes and in international collaborative work on colloids.
- Confidence can be built in regulatory assessment programmes by the conduct of independent calculations; regulators should consider the conduct of colloid transport calculations and the necessity of incorporating colloidal effects in PA.

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Appendix A

Treatment of Colloids in Recent Performance Assessments in Finland, Japan and the UK

This Appendix summarises the treatment of colloids in recent performance assessments (PAs) conducted in Finland, Japan and the UK. The treatment of colloids in SKB's PAs, and in the most recent PA for Yucca Mountain in the US is summarised in Section 3 of the main report. The treatment of colloids in other PAs not covered in this report is summarised in Bennett *et al.* (1999).

A1 Finland

Finland currently has two nuclear power plants at two sites: Olkiluoto and Loviisa. The site at Olkiluoto is owned by Teollisuuden Voima Oy (TVO). The site at Loviisa is owned by Imatran Voima Oy (IVO). The four plants at these sites together generate around one third of the electricity consumed in Finland and generate some 70 tonnes of spent nuclear fuel per year. The Finnish spent fuel disposal concept envisages burial of the spent fuel in a repository excavated at a depth of around 500 m in crystalline bedrock.

In 1983, TVO instigated a programme to select a potential deep geological disposal site for spent fuel in Finland. Each of the phases of site investigation was supported by a major PA exercise. The results of preliminary site investigations, and technical plans for encapsulation and disposal, were published in 1992 (Vieno *et al.*, 1992). An interim assessment, TILA-96, was performed in 1996 (Vieno and Nordman, 1996). In 1996, TVO and IVO established a joint company, Posiva, to manage the Finnish spent fuel disposal programme. In 1999, a PA known as TILA-99 was published (Vieno and Nordman, 1999).

A1.1 TILA-96

TILA-96 (Vieno and Nordman, 1996) was based on the use of simplified and conservative, rather than realistic models. Colloids were not considered although their potential importance was noted and the following uncertainties were highlighted as areas where further work was necessary:

- Generation of colloids at interfaces: waste-buffer, buffer-rock, redox front.
- Potential transport of colloids through the bentonite, which was considered to be a good colloid filter.
- Irreversible sorption of radionuclides onto colloids.

It was noted that if sorption of radionuclides onto colloids is reversible, then the effect of colloid transport on releases would be negligible.

A1.2 TILA-99

TILA-99 focused on the normal evolution of the repository at the candidate sites, and on the potential release and transport of radionuclides from the repository into the geosphere and biosphere. TILA-99 adopted a reference disposal concept in which spent fuel assemblies are emplaced in copper-iron canisters.

In TILA-99 colloids were again treated only qualitatively (Vieno and Nordman, 1999). It was recognised that colloids may enhance or retard transport of radionuclides through the geosphere. Enhanced radionuclide transport may result because:

- Colloids may cause the effective concentration of a radionuclide to exceed its solubility limit.
- Colloids are not influenced by matrix diffusion because of their charge and size, and radionuclides attached to colloids may therefore not be subjected to matrix diffusion processes.
- Colloids tend to stay centre-stream, where the velocity is higher due to hydrodynamic forces, during flow through fractures.

As in TILA-96, TILA-99 noted that if sorption of radionuclides onto colloids is reversible, then the effect of colloid transport is negligible with regard to release. However, TILA-99 noted that there are mechanisms by which colloids may be irreversibly incorporated into growing colloids (Laaksoharju *et al.*, 1995). Filtration and entrapment may deplete the colloid population and immobilise radionuclides attached to colloids. Alternatively, it is possible that a fraction of colloids with irreversibly sorbed radionuclides could be transported through the geosphere effectively unretarded.

A2 Japan

The Japanese Nuclear Cycle Development Institute (JNC) is the main organisation responsible for research and development activities relating to high-level waste (HLW) disposal in Japan. The Japanese national policy on radioactive waste disposal is established by the Atomic Energy Commission of Japan (AECJ). In 1989 the Advisory Committee on Radioactive Waste Management of the AECJ issued guidelines emphasising that a multi-barrier system was viewed as the most appropriate approach to geological disposal in Japan. Furthermore, it advised that a preliminary PA of the multi-barrier system should be conducted for various types of geological environment in Japan, prior to the selection of a specific geological structure or rock type for further study.

In 1992, following these recommendations and guidelines, a precursor organisation to JNC, PNC, published a report on the feasibility of geological disposal in Japan based on the results of their research (First Progress Report; PNC 1992). The strategy of the PA in the First Progress Report was to focus on the long-term performance of the engineered barrier system (EBS) to provide a basis for evaluating the feasibility of geological disposal. Colloids were not considered in the Report.

JNC submitted the Second Progress Report (H12) in 2000. The Second Progress Report distinguishes several classes of uncertainty, alternative scenarios, modelling options and parameter ranges. A list of features, events and processes (FEPs) was screened, and a diagram was constructed to describe influences among the FEPs selected for inclusion in PA models. In order to provide a baseline for comparison among a number of different scenarios, a “Reference Case” was defined. The Reference Case for vitrified waste includes emplacing steel canisters surrounded by compacted bentonite in crystalline bedrock at a depth of approximately 1,000 m. The Reference Case envisaged that the host rock is saturated with reducing, high-pH groundwater. Hydrological models consider flow in the host rock, and in major water conducting features (MWCs), with a distinct set of hydrological parameter values for each.

The Japanese disposal concept envisages the use of a compacted bentonite buffer which has a structure composed of microscopic pores. Recent experimental studies using 15 nm gold colloids and micro-organisms have confirmed that colloids do not pass through compacted bentonite, and that high molecular weight organic substances are retarded or filtered (Kurosawa *et al.*, 1997). Therefore, the effects of repository-derived colloids on performance of the disposal system are considered to be insignificant, and colloids were not considered in the Reference Case. However, the effects of colloids were modelled in a “Colloid Case”.

In order to model radionuclide migration in the geosphere taking into account the effect of colloids in groundwater, the following assumptions were made:

- Sorption of colloids to fracture surfaces is conservatively ignored.

- Diffusion of colloids into the rock matrix is conservatively ignored because the diameter of colloids is relatively large and both colloids and surfaces are electronegative.
- Sorption of nuclides to colloids is linear, instantaneous and reversible.
- The colloid concentration is constant in time and space owing to dynamic equilibrium between colloid generation and sedimentation.
- No colloids are generated by erosion of buffer materials, because of slow groundwater flow.

The equation governing the transport of colloids and radionuclides bound to colloids is then:

$$\begin{aligned} & \frac{\partial}{\partial t} [C_n + C_n^c] + n \frac{\partial C_n}{\partial x} + n_c \frac{\partial C_n^c}{\partial x} - \frac{\partial}{\partial x} D_L \frac{\partial C_n}{\partial x} - \frac{\partial}{\partial x} D_L^c \frac{\partial C_n^c}{\partial x} \\ & + l_n [C_n + C_n^c] - l_{n-1} [C_{n-1} + C_{n-1}^c] + \frac{F}{b} D_e^m \frac{\partial C_n^m}{\partial w} \Big|_{w=0} = 0 \end{aligned} \quad (\text{A.1})$$

where:

superscripts m and c denote matrix and colloid, respectively

subscripts n and c denote radionuclide and colloid, respectively

C_n = the nuclide concentration in the fractures [kg/m³]

C_n^m = the nuclide concentration in the matrix [kg/m³]

C_n^c = the nuclide concentration sorbed to colloids [kg/m³]

b = the fracture aperture half-width [m]

n = the groundwater flow rate in fractures [m/s]

n_c = the advection velocity of colloids [m]

D_L = the dispersion coefficient of radionuclides in fractures [m²/s] (= $a_L n + D_0$)

D_L^c = the dispersion coefficient of colloids [m²/s]

l_n = the decay constant [1/s]

F = the ratio of fracture area contributing to matrix diffusion [dimensionless]

D_e^m = the effective diffusion coefficient in the matrix (m²/s) (= $q^m D^m$)

x = the migration distance [m]

w = the perpendicular distance into the matrix from the fracture surface [m]

t = time [s]

Retardation due to sorption on fracture surfaces was not considered and R_n was set to 1.

The concentration of nuclides sorbed to colloids was described using equation (A.2) by assuming that sorption is linear, instantaneous and reversible:

$$C_n^c = cK_c C_n \quad (\text{A.2})$$

where:

x = the colloid concentration [kg/m³]
 K_c = the distribution coefficient of nuclides to colloids [m³/kg]

By assuming that the colloid dispersion coefficient, D_L^c , was equal to the nuclide dispersion coefficient, D_L , equation (1) can be rewritten:

$$R'_n \frac{\partial C_n}{\partial t} + n' \frac{\partial C_n}{\partial x} - \frac{\partial}{\partial x} D'_L \frac{\partial C_n}{\partial x} + R'_n |_{n} C_n - R'_{n-1} |_{n-1} C_{n-1} + \frac{F}{b} D_c^m \frac{\partial C_n^m}{\partial V} \Big|_{v=0} = 0 \quad (\text{A.3})$$

where:

$$\begin{aligned}
 R'_n &= 1 + cK_c \\
 n' &= n \left(1 + \frac{n_c}{n} cK_c \right) \\
 D'_L &= D_L \left(1 + \frac{n_c}{n} cK_c \right)
 \end{aligned} \quad (\text{A.4})$$

where R'_n , n' and D'_L were defined as the radionuclide effective retardation coefficient, effective fracture flow rate, and the effective dispersion coefficient, respectively. The influence of colloid transport was assessed by applying these effective parameters to the Reference Case. Key parameter values for both the Reference Case and the Colloid Case are provided in Table A.1.

In calculations, the following assumptions were made:

- The velocity of colloids, v_c , was taken to be the same as the groundwater flow rate, \bar{v} .
- The colloid concentration was taken to be 1 ppm (10⁻³ kg/m³), based on groundwater measurements in the Tono Mine that indicated colloid concentrations between 1 and 5 ppm, and consideration of deep groundwater colloid concentrations that have been measured at other locations worldwide.
- Colloid-radionuclide distribution coefficients were based on sorption to bentonite.

Results

The maximum dose equivalent calculated by taking into account colloid transport was several times larger than that for the Reference Case, mainly because the groundwater flux through the excavation disturbed zone (EDZ) was twice as high. The remainder of the difference occurred because colloids did not undergo matrix diffusion, and therefore radionuclides sorbed to colloids were not as effectively retarded as in the Reference Case. This effect is illustrated in Figure A.1, which shows the calculated dose rate for releases from a fault in the Colloid Case (upper diagram) and the Reference Case (lower diagram).

Parameter	Reference Case	Colloid Case
Colloid concentration	Not applicable	1ppm (10^{-3}kg/m^3)
K_c	Not applicable	1000 m^3/kg
Groundwater EDZ flux	0.001 m^3/yr	0.002 m^3/yr
Retardation factor due to sorption on fracture surface	1.0	2.0 *
Groundwater velocity	In host rock: 0,05 m/yr In MWCF: 50 m/yr	Two times larger than Reference Case

Table A.1: Key parameter values used in the analysis of colloid-facilitated transport.

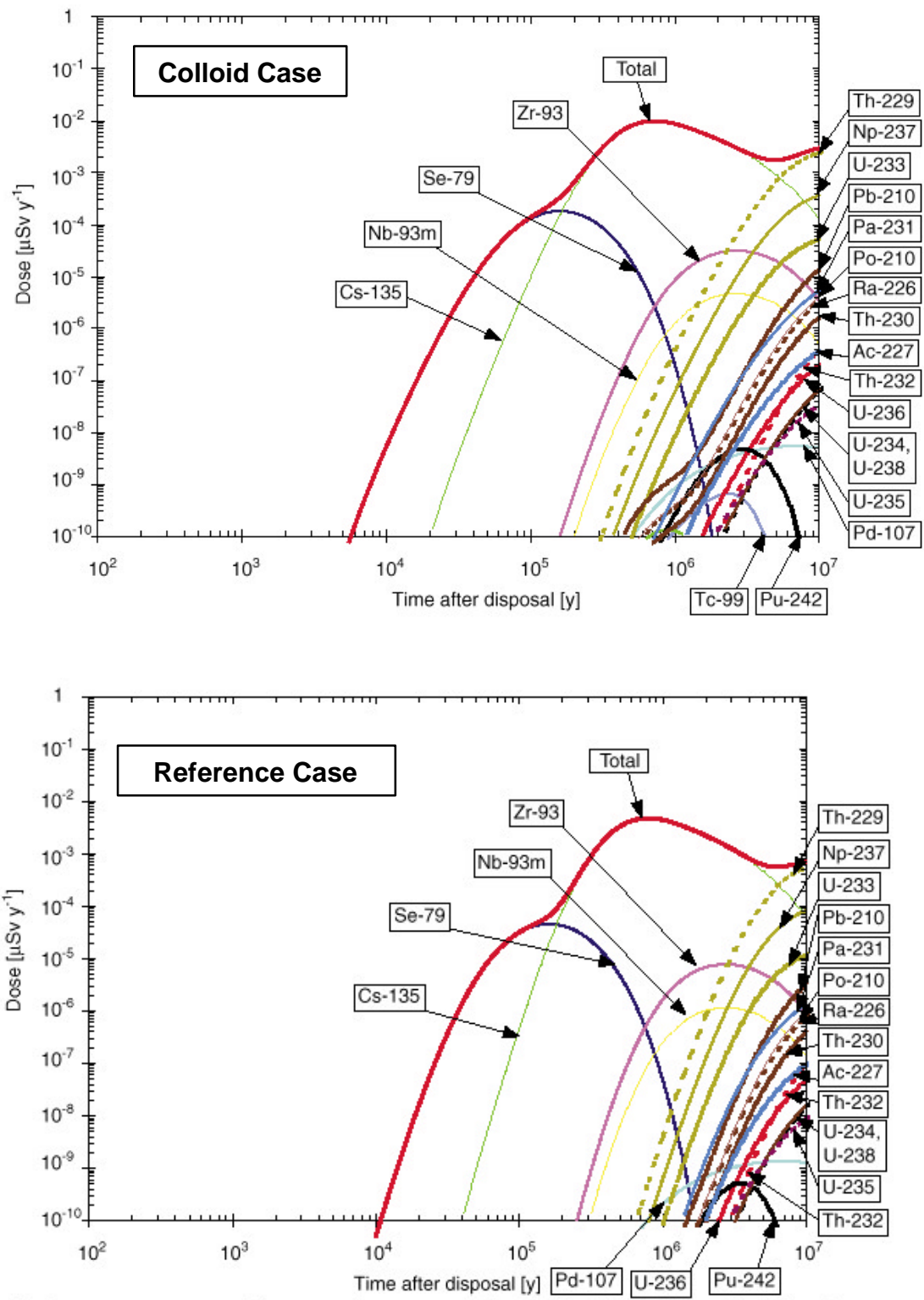


Figure A.1: Comparison of calculated dose rates including (top) and excluding (bottom) the effects of colloids (from JNC, 2000). Doses correspond to the release rate from the MWCF to the biosphere for 40,000 waste packages.

A3 United Kingdom

Nirex 97 was carried out between April 1996 and August 1997 and is a PA of a hypothetical repository for ILW and LLW at Sellafield, Cumbria (Nirex, 1997).

Nirex 97 demonstrates the approach developed by Nirex to undertake a detailed assessment programme for a candidate site. The conceptual model of groundwater flow formed the basis for numerical modelling as part of the Nirex 97 PA. Detailed models were employed for regional scale groundwater flow and transport calculations in two and three dimensions. These studies were complemented by discrete fracture network models of the repository and the surrounding host rock. These detailed models provided information concerning the pathways taken by radionuclides transported from the repository through the geosphere to the biosphere.

It was anticipated that colloid-facilitated transport would have most impact on radionuclides with low solubilities in the near-field and a strong tendency to sorb, such as the actinides (Gardiner *et al.*, 1999). Two deterministic calculations using the MASCOT PA code were made to scope the impact of colloids on risk estimates:

- In the first calculation, the effects of colloids were ‘added’ to a model in which cellulose is assumed to degrade completely, and anions and complexes are excluded from consideration. Organically-complexed radionuclides were assumed not to be retarded by matrix diffusion because of anion exclusion processes.
- In the second calculation, the effects of colloids were added to a model in which it was assumed that no organic complexants were present. Cellulose degradation products were assumed to have no effect on near-field or far-field chemical processes.

Parameter values for use in the model were selected on the basis of expert judgement. Nirex noted that within the range of uncertainty, calculations could have been undertaken that were substantially more pessimistic or optimistic than those undertaken in Nirex 97. The following assumptions were made to incorporate the effects of colloids into the MASCOT PA code:

- Spatially and temporally constant colloid concentrations were assumed in both the near-field and the far-field.
- The colloid populations in the near-field and far-field were assumed to be independent.
- Near-field colloids were assumed not to interact with the backfill or waste materials and to be mobile in the near-field porewater. Sorption of radionuclides to colloids was assumed to increase the effective near-field solubility limit and reduce the effective near-field sorption distribution coefficient.
- Colloids were assumed to travel at the same velocity as the groundwater.

In the backfill and the St. Bees Sandstone, colloids were assumed to be subject to exclusion from a fraction of the porosity, the exclusion factor specified being that appropriate to anions. In the Borrowdale Volcanic Group (BVG) and the Brockram, the colloids were totally excluded from the matrix porosity. The difference in the treatment of the BVG and the St. Bees Sandstone was justified on the basis of pore-throat diameter data.

The colloids in groundwater were assumed not to interact with the rock surfaces in the geosphere, so that the sorption distribution coefficient for the colloids in the geosphere was zero. Radionuclides were assumed to sorb reversibly onto the surfaces of colloids in both the near-field and the far-field, with linear sorption distribution coefficients.

If organic complexants were present, the distribution coefficients specifying sorption onto colloids were assumed to be subject to the same reduction factor as those specifying the sorption onto backfill in the near-field, and onto rock in the far-field. The assumed colloid populations were based on work on potential repository-derived and naturally occurring colloids undertaken as part of the Nirex Safety Assessment Research Programme (see, for example Chambers *et al.*, 1995).

Peak risks, the time at which peak risk was achieved, and the radionuclides that are significant contributors to risk, were calculated for the temperate-terrestrial, boreal-terrestrial and periglacial-terrestrial biosphere states. There was a negligible increase in the risk contribution from the ^{238}U decay chain in some of the biosphere states if the effects of colloids were added to the reference assessment model. When colloids were included in the calculations in which organic complexants had little impact, the risk contribution from the ^{238}U decay chain increased by a factor of three to four.

For the parameter values used, the impact of organic complexants is much greater than that of the colloids, so it is only in models in which organic complexants have little impact that the risk estimate may be underestimated by neglecting the effects of colloids. The impact of colloids is greatest after long periods, when the risk is dominated by the contribution from the ^{238}U decay chain. It should be noted that at the late times at which the ^{238}U decay chain dominates the risk estimate, there are many other uncertainties. In order to strengthen the finding that the effect of colloidal transport of radionuclides is limited, Nirex concluded that consideration should be given in future to a wider region of parameter space and to the validity of the assumptions in the approach.