WBS: 1.2.5.4
QA: L

Civilian Radioactive Waste Management System
Management & Operating Contractor

Chapter 7
Total System Performance Assessment-Viability Assessment (TSPA-VA) Analyses Technical Basis Document

Unsaturated Zone Radionuclide Transport

B00000000-01717-4301-00007 REV 00

August 14, 1998

Prepared for:

U.S. Department of Energy
Yucca Mountain Site Characterization Office
P.O. Box 30307
North Las Vegas, Nevada 89036-0307

Prepared by:

TRW Environmental Safety Systems Inc.
1261 Town Center Drive
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DE-AC08-91RW00134

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August 14, 1998

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ACKNOWLEDGMENT

Unsaturated Zone Radionuclide Transport

The development of the Total-System Performance Assessment-Viability Assessment Technical Basis Document has benefited from the support of people from many different organizations and disciplines from across the Yucca Mountain Project.

The following persons provided support in developing the technical basis of various components of the TBD:

- 3-D UZ Transport Model Development and Sensitivity Studies: Bruce Robinson and Chunhong Li
- Sorption, Diffusion, and Colloid Parameter Development: Ines Triay, Arend Meijer, Christine Stockman, and David Sassani
- Radioactive Decay Process Model and Abstraction: Chunhong Li and Anthony Smith
- Coupling with Total System Performance Assessment Model (RIP): Bruce Robinson, Chunhong Li, Vinod Vallikat, Patrick Mattie, David Sevougian
- Transient Flow and Transport Sensitivity Study: Andrew Wolfsberg
- Sensitivity Studies on Linear Sorption Models and Mineralogic Heterogeneity, Hari Viswanathan and Bruce Robinson
- FEHM Model Gridding: Carl Gable

The clarity and technical completeness of the work in this document has been greatly improved by the careful and thoughtful technical reviews and subsequent discussions provided by:

Steve Alcorn , Robert Baca, Tom Doe, Ian Miller, Mark Nutt, Mark Tynan, Eric Zwahlen, PA Peer Review Panel.

The transparency and traceability of the data and supporting references were ensured by the support of the following individuals:

Carol Crawford, Patricia Gibson, Steve Harris, Robert Howard, Celister Houston, Raymond Keeler, Orville Pratt, Robert Sessions, Kathie Sinclair, Mary Stuart

The ability to convey the specific conceptualizations and the results of this very complex set of analyses and documentation has been enhanced due the contributions of :

Kathy Gaither, Lin Henderson, and RED, Inc.

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ACRONYMS

1-D	one-dimensional
2-D	two-dimensional
3-D	three-dimensional
CEC	cation exchange capacity
CFu	Crater Flat undifferentiated
CHn	Calico Hills nonwelded
DKM	dual permeability model
DTN	data tracking number
ECM	equivalent continuum model
ESF	Exploratory Studies Facility
FEHM	Finite Element Heat and Mass
LANL	Los Alamos National Laboratory
LBNL	Lawrence Berkeley National Laboratory
LTA	long-term average
MTU	metric tons uranium
NSP	Nevada State Plane
Np	neptunium
PDV	primary dependent variable
PTn	Paintbrush nonwelded
RTD	residence time distribution
RTTF	residence time/transfer function
SDV	secondary dependent variable
SZ	saturated zone

Tc	technetium
TCw	Tiva Canyon welded
TOSPAC	Total System Performance Assessment Code
TSPA	VATotal System Performance Assessment-Validity Assessment
TSw	Topopah Spring welded
USGS	United States Geological Survey
UZ	unsaturated zone

7. UNSATURATED ZONE RADIONUCLIDE TRANSPORT

7.1 INTRODUCTION

The repository, if constructed, will be sited in the unsaturated zone (UZ) at Yucca Mountain, located approximately 350 m above the water table and roughly 200 to 300 m below ground surface, depending on the local topography. In the UZ, available voids in the rock (rock matrix pores and fractures) are filled partially with water and partially with air. The regional water table is the top of the saturated zone (SZ), below which the rock voids are fully saturated with water. Water movement is primarily downward through the UZ at Yucca Mountain, with possible transverse flow in certain regions such as perched water.

Under nominal and most disruptive scenarios, the transport of radionuclides through the UZ is one of several processes that impede the release of radioactive waste to the SZ after repository closure. Radionuclides released from the potential repository may enter the UZ and be carried by the natural downward movement of water to the SZ. The radioactive waste may migrate in UZ groundwater as a dissolved molecular species or associated with particles called colloids. Colloids are fine particles (between 0.001 and 1 micron) that can interact with radionuclides through sorption and other chemical mechanisms, and are themselves potentially mobile. The sequence of hydrogeologic units, their hydrologic and geochemical characteristics, and the imposed infiltration flux control radionuclide transport in the UZ.

7.1.1 Hydrogeologic Overview

Yucca Mountain was formed from a sequence of volcanic ash flows and ash falls that created a lithostratigraphic sequence of welded and nonwelded tuffs (Sawyer et al. 1994), as shown in Figure 7-1. The lithostratigraphic sequence of the UZ of Yucca Mountain is composed of alternating layers of welded and nonwelded volcanic tuff that are tilted, uplifted, fractured, and faulted. The welded tuffs, formed by massive ash flows, are dense, brittle rocks with relatively low matrix porosity and a high fracture density. The nonwelded tuffs, formed from smaller ash flows and ash falls, are a more porous and elastic rock having relatively high matrix porosity and lower fracture density. The nonwelded tuffs can have either high or low matrix permeabilities depending primarily on the degree of alteration of the rock minerals into zeolites. Zeolitic units are common in the lower portions of the UZ and have matrix permeabilities similar to those in the welded tuffs. Nonwelded tuffs that have not been altered to zeolite, such as the nonwelded vitric tuffs, have much larger matrix permeabilities. Hydrogeologic stratigraphy of the UZ at Yucca Mountain is illustrated in Figure 7-1. The Calico Hills nonwelded (CHn) unit may be further divided into vitric and zeolitic units.

From the surface, the highly fractured Tiva Canyon welded (TCw) hydrogeologic unit is the first major hydrogeologic unit encountered under a largely unconsolidated alluvial cover that varies in thickness from 0 m to more than 50 m. Below the TCw is the Paintbrush nonwelded (PTn) unit, with a higher permeability in the rock matrix, but lower bulk permeability due to lower fracture density (Bodvarrson et al. 1997). The potential repository is currently sited within the middle nonlithophysal, lower lithophysal, and lower nonlithophysal portions of the Topopah Spring welded (TSw) unit. This thick-welded unit consists mainly of highly welded and fractured tuff

subunits, some of which contain a high density of lithophysal cavities. The TSw extends about 100 to 150 m below the potential repository before the CHn unit is reached. The CHn is a less fractured tuff that contains both vitric sections and a horizon that exhibits extensive zeolitic alteration (Bodvarrson et al. 1997; Chipera et al. 1997a,b). In the southwest portion of the conceptual repository block, the underlying Crater Flat undifferentiated (CFu) unit also lies above the water table, and has zones of zeolitic alteration (Chipera et al. 1997a,b). In general, the major hydrogeologic units dip to the East at angles of 5 to 10 degrees, creating the possibility of lateral flow at the interfaces of units of contrasting hydrologic properties (Bodvarrson et al. 1997). The predominant modes of fracturing are cooling joints and fractures and faults induced by past tectonic activity. Experimental laboratory and field measurements, as well as conceptual and numerical models (Montazer and Wilson 1984; Wittwer et al. 1995; Altman et al. 1996; Robinson et al. 1996), suggest that some of these features may be associated with complex subsurface flow patterns.

An important characteristic of both the welded tuffs and nonwelded zeolitic tuffs is the difference in bulk permeability between the fractures and the matrix. The fractures typically have bulk permeabilities that are several orders of magnitude larger than the matrix permeabilities, and fracture porosities that are orders of magnitude smaller than the matrix porosities. The differences are important characteristics of these tuffs because the high-permeability, low porosity fractures can provide a much more rapid transport pathway to the water table than the matrix. On the other hand, the unaltered (nonwelded) tuffs have larger matrix permeabilities that are of the same order of magnitude as the fracture permeabilities. These tuffs play an important role in the hydrologic behavior of the UZ because fracture flow is strongly attenuated, transport velocities are much slower, and contact of dissolved materials with the rock matrix is enhanced.

Perched water has been observed in some boreholes penetrating the repository, mainly in the northern area of the repository and along the western border of the Ghost Dance Fault (see Figure 7-1). Perched water is a localized SZ that lies above both unsaturated rock and the regional water table and typically forms on top of a relatively impermeable hydrogeologic unit. When present, perched water is typically found at the base of the TSw and in areas of the Calico Hills zeolitic unit. These perched water bodies are believed to have formed by alteration of minerals in fractures to low permeability minerals that effectively block flow through the fractures (Bodvarsson et al. 1997, Section 13.5). This blockage of fracture flow will lead to lateral diversion of radionuclide transport if the percolation flux is sufficiently large.

7.1.2 Conceptual Overview of Flow and Transport in the Unsaturated Zone at Yucca Mountain

Our present conceptual model for the hydrochemical system at Yucca Mountain has evolved from the earlier hydrologic models of Winograd (1981) and Montazer and Wilson (1984) and more recent hydrochemical data (Fabryka-Martin et al. 1996b-c; Flint et al. 1996; Bodvarsson et al. 1997; Levy et al. 1997; Yang et al. 1996, 1997), and is the product of the integration of hydrologic, mineralogic, structural, hydrochemical and geochemical field and laboratory data collected and used to characterize transport processes in the UZ at Yucca Mountain. This conceptual model is discussed below relative to the perceived behaviors for low and high infiltration rates, representing modern and ancient climates (see Section 2.2), respectively.

TCw—Surface infiltration - Under all climatic conditions the downward migration of meteoric waters through the TCw occurs via fracture flow. The water chemistry will reflect spatial and temporal changes in evapotranspiration and is not expected to undergo any modifications during the rapid downward migration of surface waters through the TCw, to the top of the PTn (Robinson et al. 1997).

PTn—Transition from fracture dominated flow to matrix dominated flow - Under low infiltration rates and with the exception of discrete fracture flow paths, associated with large scale faults at the surface (Levy et al. 1997), matrix flow is the dominant mechanism sustained throughout the PTn. For higher infiltration rates, fracture flow is more predominant but only represents approximately 20 percent of the total water flux (Robinson et al. 1997; Chapter 6). This is because these discrete "fast paths" are not ubiquitous, are not associated with large catchment areas at the surface, and therefore do not involve large volumes of water. Under present climate conditions, fractures do not contribute substantially to the total water flux through the PTn even though they may represent "fast pathways" (Robinson et al. 1997). The water chemistry in the PTn matrix pore waters is believed to represent a mixture of modern meteoric waters derived directly from the surface, and having undergone different degrees of evapotranspiration.

TSw—Transition from matrix dominated flow to fracture dominated flow - Under all expected climatic conditions, the flow of water in the TSw is predominantly via fracture flow. As in the case of the TCw, ground water travel times from the base of the PTn to the top of the basal TSw vitrophyre are short in comparison with travel times through units with matrix dominated flow such as the PTn. In terms of water chemistry, different degrees of chemical maturation may occur as a function of infiltration rate, as indicated by the solution- precipitation textures from Yucca Mountain fracture fillings (Vaniman and Chipera 1996). At high infiltration rates, a substantial component of flow down-dip along the top of the basal TSw vitrophyre is possible in many areas below the potential repository. These areas of lateral diversion are also likely locations for the formation of perched water bodies. This lateral flow combines with a vertical flow component to produce a mixed perched water chemistry with low chloride concentrations at the top of the vitrophyre. Relatively slow seepage from perched water bodies and/or during lateral flow is expected through fractures in the basal TSw vitrophyre. These seepage waters possibly mix with underlying waters into the underlying nonwelded tuffs of the CHn hydrogeologic unit. At low infiltration or in the case where the vitrophyre does not appear to be a barrier to vertical flow locally, water flux is unimpeded and reaches the top of the CHn hydrogeologic unit.

CHn—Composite vertical flow and lateral diversion - The Calico Hills nonwelded tuffs are composed of interlayered zeolitized and non-zeolitized units underlying the repository horizon. Slow percolation through intermingled matrix and fractures pathways in vitric and zeolitic rocks, is expected to retard and divert vertical flow through the mountain. In all cases, the flow in the CHn is complex due to the complex spatial distribution of vitric and zeolitic rocks (Robinson et al. 1997; Chapters 6 and 12). The perched water chemistries should reflect mixing as in the case of the basal TSw vitrophyre (Robinson et al. 1997). In the case where the basal vitrophyre is non-existent or bypassed by fracture flow, a permeability barrier may occur, due to the low permeability of zeolitic rocks (when present) in the CHn hydrogeologic unit. This mechanism is possible for any infiltration rate and can result in the formation of perched water and lateral flow

within the vitric CHn. The pore water chemistry is expected to be strongly influenced by both the lateral flow component within the matrix (i.e., high chloride concentrations) and the vertical flow beneath the perched water bodies (i.e., low chloride concentrations).

Water Table—Seepage from the base of the CHn into the saturated zone - This conceptual model allows for some fast pathways through faults that cut the original stratigraphic sequence, but under present climatic conditions, these pathways are not associated with the bulk of the water flux through the mountain. Both the PTn and the TSw Basal vitrophyre/CHn horizons play an important role in retarding the flow of meteoric waters to the water table.

Fault Zones—Potential fast pathways - Evidence of $^{36}Cl/Cl$ ratios, with a source attributed to nuclear bomb testing roughly 50 years ago, has been found in some sections of the Exploratory Studies Facility (ESF) that lie in the TSw hydrogeologic unit. The only available transport pathway for the "bomb-pulse" $^{36}Cl/Cl$ ratio to penetrate to the TSw is by migration from the surface as dissolved ions in the infiltrating water. The identification of this bomb-pulse isotopic signal in the ESF indicates that some transport pathways either bypass the PTn matrix or are regions of highly focused infiltration through the PTn. Travel times for the bomb-pulse ^{36}Cl, under conditions of uniform infiltration through the PTn matrix, would require hundreds to thousands of years to penetrate the PTn. Levy et al. (1997) extend the work of Fabryka-Martin (1996b) and provide an analysis of structural controls of fast pathways from ground surface to the ESF. They note that the locations at which these fast pathways have been identified in the ESF appear distinctive, with the primary controls on their distribution being the presence of faults that cut the PTn, the magnitude of surface infiltration, and structural features that result in lateral diversion of flow away from fault zones.

Transport Characteristics - Transport from a potential repository source is affected by the sorptive interactions with the rock and the degree of contact between radionuclides and the rock matrix. Some radionuclides, such as ^{99}Tc, do not sorb. Other radionuclides, such as ^{237}Np, move at a slower rate than a nonreactive tracer due to moderate sorptive interaction with various rock types. Still others, such as aqueous ^{242}Pu, are found to strongly interact with all rock and, therefore, are relatively immobile. Nevertheless, sorptive interaction is only one part of the mechanism needed to retard the movement of radionuclides. Radionuclides that are transported through fractures cannot sorb onto the rock matrix without some mechanism that allows the radionuclides to contact the rock matrix. (Although sorption onto minerals along fracture surfaces is likely, difficulty in characterization of this sorption mechanism has lead to the conservative assumption used for TSPA-VA of no sorption in the fractures.) For example, many radionuclides have been found to strongly sorb to zeolitic rock. However, highly zeolitized rock generally has low matrix permeability, and in some cases, low fracture permeability as well. The low-permeability character will lead to transport pathways that bypass the zeolitic minerals, due to lateral diversion or transport through fractures, severely limiting the degree of contact between radionuclides and zeolitic minerals. Therefore, low levels of zeolitic alteration in the CHn vitric, which do not severely reduce matrix permeability, are found to have more influence on the transport of sorbing radionuclides. The effects of lateral diversion and focused flow in certain regions of the potential repository also tend to reduce the degree of contact between radionuclides and both the CHn vitric and zeolitic rocks. The more evenly distributed percolation flux in the TSw allows for more intimate contact of radionuclides with rock matrix, through matrix diffusion and advection, despite the low matrix permeabilities of the TSw.

Sorptive interactions may enhance radionuclide transport if the aqueous species sorbs to mobile colloids. Colloid-facilitated transport enhances the movement of the aqueous species because the sorptive interaction with matrix is reduced (hence reducing retardation in transport) and colloids may tend to move preferentially through the higher-velocity fracture pathways. In addition to reversible, sorptive type interactions with colloids, radionuclides may also be irreversibly attached to colloids (e.g., coprecipitation during colloid formation). Isotopes of Pu have been identified as radionuclides that are likely to be affected by colloid-facilitated radionuclide transport (see Sections 6.4.2 and 6.5).

Radionuclides that have little or no sorptive interaction with the welded tuff matrix are expected to migrate through the TSw relatively quickly due to advective transport through fractures. The nonsorbing radionuclides travel primarily through fractures except for transport through the CHn vitric, where matrix flow and transport is expected to dominate. Therefore, transport times from the potential repository to the water table for nonsorbing radionuclides such as ^{99}Tc, and ^{129}I are primarily governed by transport in the CHn vitric.

Another factor that affects travel time to the water table is the lateral diversion of flow above the CHn. Although the lateral diversion increases the transport path length to the water table, the transport pathways are primarily fracture pathways. The diverted percolating flow eventually finds some pathway to the water table. Therefore, lateral diversion can lead to zones of focused flow to the water table, where the flow rates may be locally magnified well beyond the flow rates anticipated for uniform vertical percolation. The travel times for radionuclides transported through a focused percolation zone will tend to be relatively short, including transport through the CHn vitric. This section (transport characteristics) is a summary of some of the qualitative findings from work documented in this chapter.

7.1.3 Interfaces to Other Processes

7.1.3.1 Unsaturated Zone Flow

The separation of UZ radionuclide transport from UZ flow is a process abstraction that we use for sensitivity studies and development of the TSPA-VA UZ radionuclide transport model. We combine the two processes in TSPA-VA calculations by using a set of pre-calculated, three-dimensional (3-D), UZ flow fields, computed with the site-scale UZ flow model. These flow fields are incorporated directly into the 3-D, UZ radionuclide transport model for TSPA-VA.

7.1.3.2 Thermal Hydrology

The waste heat released in the potential repository influences UZ flow and temperature behavior. As for UZ flow, the separation of thermal hydrology and UZ radionuclide transport is a process abstraction. If we may assume that the thermal-hydrologic effects of the repository do not result in any permanent changes to the mineralogic or hydrogeologic conditions of the UZ, then the effects of the thermal-hydrologic processes on radionuclide transport are believed to be minor. Our models of this process (see Section 7.6.3) indicate that the time period during which the temperature and flow fields are significantly perturbed occurs prior to the release of most of the radionuclides. Therefore, the thermal-hydrologic effects on the UZ temperature and flow fields are assumed to have a negligible effect on radionuclide transport. However, the effects of

thermal-hydrology are still important for defining the behavior of the waste package and radionuclide releases from the engineered barrier system. Therefore, thermal-hydrology is included for these subsystem models (see Chapters 5 and 6).

The effects of thermal perturbations can also potentially have long-term consequences relative to minerals in the UZ and change both hydrogeologic and transport properties of the system. These types of thermal-hydrologic changes to the system may affect long-term radionuclide transport in the UZ. Although our present evaluation of these coupled processes is not complete, we have conducted TSPA sensitivity calculations concerning off-normal behavior to address this coupling (see Chapter 11).

7.1.3.3 Waste Form Mobilization

The waste form mobilization process provides the radionuclide fluxes at the emplacement drift boundary for UZ radionuclide transport calculations. The emplacement drift boundary represents a spatial-domain abstraction interface between processes that affect radionuclide transport inside the emplacement drift and radionuclide transport in the UZ. Therefore, the radionuclide fluxes calculated at the emplacement drift wall are a source term for UZ radionuclide transport calculations. This source term is expected to provide radionuclide fluxes at the emplacement drift boundary that will vary as a function of the location in the potential repository and time.

7.1.3.4 Saturated Zone Radionuclide Transport

SZ flow and radionuclide transport calculations use the results of the UZ radionuclide transport calculation to assess the radionuclide migration to the accessible environment. The water table in the vicinity of the potential repository represents a spatial-domain abstraction interface between processes that affect radionuclide transport in the unsaturated and SZs. The radionuclide fluxes calculated at the water table due to UZ radionuclide transport are a source term for SZ radionuclide calculations. This source term is expected to provide radionuclide fluxes at the water table that will vary as a function of position on the water table and time. Although no feedback mechanisms have been identified between SZ radionuclide transport and UZ radionuclide transport, the position of the water table is recognized as a feedback from SZ flow to UZ radionuclide flow and transport.

7.1.4 Previous Treatments in YMP PA Modeling Efforts

7.1.4.1 TSPA-1995

Unsaturated-zone transport has been dealt with in a number of ways in past TSPAs. In TSPA-1995 (CRWMS M&O 1995a), the Markovian algorithm in the RIP TSPA code was used (Golder 1994). This algorithm assumes a random transitioning of radionuclide particles between fractures and matrix according to user-specified Poisson-process transition rate. Within either the fracture or matrix flow mode, the particles move by plug flow and may be retarded according to an equilibrium K_d sorption model. Velocities and fluxes in both the fractures and matrix were derived from the site process-level hydrology model using the TOUGH2 (Transport of Unsaturated Groundwater and Heat) code (Wittwer et al. 1995; Bodvarsson and Bandurraga

1996). Exact details of the TSPA-1995 UZ transport model are given in Section 7.4 of CRWMS M&O (1995a). Since the RIP transport model is more of a descriptive model than a process-based model, the validity of the Markovian transport algorithm must be established by comparing breakthrough times from the RIP code over a range of parameter and model uncertainties to breakthrough times (to the water table) from the 3-D site-scale UZ transport model (Robinson et al. 1995; Robinson et al. 1996).

The RIP4.05a model used for TSPA-1995 is based on a phenomenological approach that attempts to describe rather than explain the transport system. The resulting transport algorithm is based on a network of user-defined *pathways*. The pathways may be used for both flow balance and radionuclide transport purposes, and may account for either gas or liquid phase transport. The purpose of a pathway is to represent large-scale variations of the hydrologic system, such as geologic structures and formation-scale hydrostratigraphy. Fracture/matrix exchange at stratigraphic unit boundaries is also modeled in RIP405a, and is based on the total proportion of the flow that is flowing within each of the two downstream flow modes. RIP405a does not contain an abstraction for exchange due to matrix diffusion. Dispersion is not explicitly modeled in the dual flow-mode model, however the fracture/matrix interaction process does cause longitudinal dispersion of radionuclides (which may or may not follow a Fickian description). Linear sorption for radionuclide matrix or fracture transport may be included in RIP calculations by adjusting the radionuclide velocities according to the appropriate retardation factor. For TSPA-1995 (CRWMS M&O 1995a), retardation factors were employed for certain radionuclides and rock types (different stratigraphic layers) for matrix transport, but radionuclides were assumed to not sorb during fracture transport.

The TSPA-1995 calculations performed with RIP assumed one-dimensional (1-D) transport through a set of six columns. Five stratigraphic layers were used to represent the variations in rock type between the potential repository and the water table. These variations were based primarily on the degree of welding found in the volcanic tuffs that comprise the UZ. The six columns were used to approximate the areal variations in stratigraphy and distance to the water table throughout the potential repository. A separate UZ steady flow model abstraction was used to define the steady flow in fractures and matrix between the potential repository and the water table (CRWMS M&O 1995a).

The assumption of 1-D transport in RIP was investigated for a limited set of conditions. Calculations of arrival time at the water table using the TOUGH2 unsaturated flow process model and a detailed two-dimensional (2-D) cross-section from the United States Geological Survey (USGS) site-scale model were compared with stochastic RIP calculations to investigate the 1-D approximation used in RIP (CRWMS M&O 1995a). The results of this comparison indicated that the range of results obtained from 1-D transport calculations in RIP using stochastic sampling of hydrogeologic and transport properties and infiltration rates bracketed the results found by the process model.

The intra-unit fracture matrix model in RIP is based on a random Markov process for transitioning particles between the fracture and matrix continua. The primary model parameter is an empirical interaction parameter called the Poisson transition rate. To help set this parameter, comparison calculations between RIP and Finite Element Heat and Mass (FEHM) were conducted. FEHM (Robinson et al. 1997) is a process-level model that solves the basic

differential conservation equations for unsaturated flow and transport. The conservation equations used by FEHM describe physical and chemical mechanisms that affect transport with a dual-continuum option for modeling fractured, porous rock. Chemical interactions are limited to a selection of different equilibrium adsorption models. FEHM has been used to qualitatively calibrate the fracture/matrix exchange coefficient in RIP (CRWMS M&O 1995a). A sensitivity study was also performed using FEHM that considered the relation of the fracture/matrix exchange coefficient in RIP and the more fundamental physical processes captured in FEHM (CRWMS M&O 1995b). Although matrix diffusion was not modeled in TSPA-1995, simulations with FEHM indicated that matrix diffusion can greatly delay transport through the UZ (CRWMS M&O 1995b). For TSPA-VA, there are no plans to used the RIP Markov-process transport model.

The principal differences for the UZ radionuclide transport model in TSPA-VA as compared with the TSPA-1995 and TSPA-1993 are the much greater infiltration flux in TSPA-VA and the 3D flow model including perched water. These features have resulted in more rapid radionuclide transport to the water table. Both of these items are described in Chapter 2, but briefly the consequences for UZ transport may be summarized as follows.

- Infiltration flux rose from an average value of 1.25 mm/yr for the "high infiltration" present day climate (3.75 mm/yr for long-term climate conditions) in TSPA-1995 to 7.7 mm/yr for present day and a long-term average (LTA) climate infiltration rate of 42 mm/yr in TSPA-VA. This large increase in infiltration has resulted in flow fields that are now dominated by fracture flow in the welded units for all climate conditions.

- Perched water was not modeled in previous TSPAs. Perched water has been included in the flow model for TSPA-VA by reducing permeability in the fractures of units below locations where perched water is believed to exist. The locations are generally at or near the TSw/CHn interface and, therefore, lie between the potential repository and the water table, primarily in the north-eastern portion of the potential repository block. This has resulted in large-scale diversion of radionuclide transport around these low-permeability units. The diversion flow is primarily through fractures and results in focused flow to the water table around the perched water. Therefore, the lateral diversion has resulted in more rapid transport to the water table than for more 1-D, vertical transport pathways. Previous TSPAs assumed the transport pathways to be 1-D.

7.1.4.2 TSPA-1993

In one TSPA-1993 (Andrews et al. 1994), the Markovian algorithm was also used. In the other TSPA-1993 (Wilson et al. 1994), as well as TSPA-1991 (Barnard et al. 1992), a 1-D, dual-continuum model of solute transport was used. The model is implemented in the TRANS module of the computer program Total System Performance Assessment Code (TOSPAC) and is described in detail in Dudley et al. (1988) and Gauthier et al. (1992). TRANS was used to calculate transport in both the UZ and SZ for the equivalent continuum model (ECM)-flow cases. With the WEEPS model, TRANS was only used in the SZ; transport in the UZ was assumed to be rapid and complete, and the source output was introduced directly into the SZ. WEEPS is a model abstraction used to represent fracture-only flow and transport.

TRANS solves two generalized advection-dispersion equations simultaneously, one for solute transport in the matrix, the other for transport in the fractures. The equations include an advective term, a term for diffusion and hydrodynamic dispersion, a term for radionuclide decay and production, a source term, and a term specifying transfer between the matrix and fracture continua. The equations are modified to disallow hydrodynamic dispersion upstream from an internal source region. Radionuclide decay chains can be accounted for. Transport of sorbing radionuclides is modeled using the linear K_d approximation. The transfer term describes both advective and diffusive coupling between the matrix and fractures. The transfer term also includes a factor to represent the matrix/fracture coupling strength (for example, to mimic fracture coatings), but this factor has only been varied in recent calculations, and not in TSPA-1991 or TSPA-1993. TRANS calculates the concentration of each radionuclide in time and space, the cumulative amount of each radionuclide that crosses each boundary, and the dose that would result from drinking the water at a given location.

7.1.4.3 Synopsis of Current Treatment and Changes from Prior Efforts

7.1.4.3.1 Coupled RIP / FEHM Model

As described in Section 7.8.5.1, RIP is more of a descriptive code than a process-based model. It more or less represents what has already been determined based on the detailed process-level models and codes, which in the case of the UZ would be TOUGH2 for fluid flow (Bodvarsson et al. 1997) and FEHM for flow and radionuclide transport (Robinson et al. 1996). In TSPA-1995, hydrologic simulations of UZ flow were conducted with TOUGH2, and then the resulting fracture and matrix fluxes from TOUGH2 were abstracted into RIP. The various UZ flow abstractions were of several types. For example, coarser discretization of the hydrogeologic units was used in RIP, which required some averaging of the greater number of TOUGH2 hydrogeologic units. Also, in TSPA-1995, many TOUGH2 runs were conducted as a function of uncertainty in the flow parameters. The results of these multiple runs were represented as a "wide band distribution" of matrix/fracture fluxes and velocities as a function of the uncertain infiltration rate. (The spread in the wide-band distribution at a given infiltration rate was due to property uncertainty and heterogeneity—see CRWMS M&O 1995a.) This wide-band distribution was sampled probabilistically (uniformly) in RIP.

Also, as already mentioned, the transport in TSPA-1995 was represented using the RIP Markovian algorithm for interaction between matrix and fractures in a given hydrogeologic unit. This is again a descriptive model rather than a process model—a descriptive model that must be "verified" against the process model, FEHM. Also, all the UZ flow and transport in TSPA-1995 was represented with a set of six parallel 1-D vertical columns that covered the repository area and went from the repository horizon to the water table. (This type of geometric/dimensionality abstraction neglects lateral flow.)

Because the above types of flow and transport abstractions are not particularly transparent (and appear to be quite removed from the process models) and because the NRC has had reservations about the Markovian approach, a different UZ transport model has been implemented for TSPA-VA. Instead of making the sorts of abstractions described above, which also require extensive post-processing of process-model results (e.g., TOUGH2 flow fields) in order to input them into RIP pathways, the FEHM particle tracker has been coupled directly to RIP. UZ transport

calculations are performed using the 3-D site-scale Los Alamos National Laboratory (LANL) transport model (FEHM particle tracker), with 3-D flow fields generated by the TOUGH2 Lawrence Berkeley National Laboratory (LBNL) site-scale flow model. This is more like the approach for TSPA-1993 (Wilson et al. 1994) which used a flow/transport process model (1-D) directly embedded into the TSPA probabilistic code.

There is really nothing wrong with the cells approach or even the Markovian pipe approach. They will run much faster, but require a bit more justification to prove that they accurately represent the physical processes. However, it is more transparent to couple the process model directly with RIP, though it requires longer computation time and more computational resources.

7.1.4.3.2 Unsaturated Zone Flow and Water Saturations

As discussed in the previous section, the 3-D site-scale transport model (FEHM particle tracking) is coupled directly with the RIP for TSPA-VA calculations of UZ radionuclide transport. The particle tracking calculations use 3-D, dual permeability, flow fields generated by the TOUGH2 UZ flow model. Variability in the flow fields generated by TOUGH2 correspond to the expected range of infiltration and flow parameter variations (see Chapter 2). These flow fields are incorporated into the particle tracking transport model through a direct translation of the fluxes between grid blocks and water saturations for both the fracture and matrix continua. This method differs from previous TSPA calculations in that the effects of three dimensional flow patterns are captured directly in the TSPA UZ radionuclide transport calculations.

7.1.4.3.3 Particle Tracking

The particle tracking technique we use for simulating UZ radionuclide transport is based on residence time/transfer function (RTTF) particle tracking technique. This method employs a cell-based approach that sends particles from node to node on a finite difference or finite element grid, after keeping each particle at the cell for a prescribed period of time. To incorporate transport mechanisms such as dispersion and matrix diffusion, the residence time of a particle at a cell is computed using a transfer function that ensures that the correct distribution of times at the cell is reproduced. The method is a significant departure from the traditional procedure of mapping a particle trajectory by resolving the velocity vector at a given position through interpolation between positions on the grid. The new procedure is computationally very efficient, enabling large-scale transport simulations of several million particles to be completed rapidly on modern workstations. Furthermore, since the cell-based approach uses directly mass flow rate information generated from a numerical fluid flow solution, complex, unstructured computational grids pose no additional complications. A number of numerical validation and example calculations are presented to demonstrate the strengths of the new technique (see Section 7.4). A discussion is also provided to point out those situations for which the assumptions of the method might lead to unacceptable numerical inaccuracy, and for these cases alternative numerical transport techniques are recommended (see Section 7.4). For TSPA-VA, the technique is used for UZ radionuclide transport simulations, since the limitations of the method are minimal for the UZ, and the cell-based approach allows accurate simulation of dual-permeability systems in which there is a vast disparity in the travel times depending on whether the transport is in the fractures or the matrix.

7.1.5 QA Status of Data and Simulations

A summary of the data tracking numbers (DTNs) for the relevant data and simulations used in this chapter is provided in Tables 7-1 and 7-2. Table 7-1 lists the source data that are used to model UZ radionuclide transport in TSPA-VA. Table 7-2 provides a summary of the software codes, input and output files, and DTNs associated with technical figures produced in this chapter. Unless otherwise noted, the codes used in this chapter are not yet qualified. Qualification status for FEHM (Finite Element Heat and Mass), which is the process-level code used in this chapter for UZ transport calculations, is referenced by Zyvoloski et al. (1997).

7.1.6 Roadmap of Chapter 7

The remainder of this section will cover the assumptions, models, parameters, and bases for the UZ radionuclide transport calculations for TSPA-VA. Concepts relating to transport mechanisms of advection under steady and transient flow conditions, diffusion, sorption and colloids are reviewed in Section 7.2. The abstraction testing workshop for UZ radionuclide transport, conducted in February of 1997, identified and prioritized issues and established analyses plans to support TSPA-VA. The results of the workshop are summarized in Section 7.3. The approach used for TSPA-VA is given in detail in Section 7.4. Here details of the particle tracking method are discussed, along with detailed discussions of process abstractions to be used with the particle tracking model. The process abstractions pertain to long-term transient flow due to climate change, colloid-facilitated radionuclide transport., and methods to treat fine-scale mineralogical heterogeneity. Details of the implementation for TSPA-VA, including coupling of the UZ radionuclide transport model with the total system performance assessment code, RIP, are given in Section 7.5. Examples of coupled RIP/FEHM calculations for the TSPA-VA base case are provided and the transport parameter ranges used for TSPA-VA are presented. Results of sensitivity tests and additional process abstractions are given in Section 7.6. Sensitivity calculations considering the effects of parameter variations for matrix and fracture sorption, matrix diffusion, and fracture/matrix flow configurations. Sensitivity calculations for Np sorption as a function of chemical environment are presented and discussed relative to the use of a linear sorption model. The effects of episodic transient flow and transport are investigated for 1-D systems.

7.2 PROCESS MODEL DESCRIPTION

7.2.1 Advection

Advection is the movement of dissolved or colloidal material because of the bulk flow of a fluid, which in this case is water. This key transport mechanism can carry radionuclides through the approximately 300 m of unsaturated rock between the potential repository and the water table. Advection is also an important mechanism for radionuclide movement between fractures and rock matrix. In many of the hydrogeologic units, advection through fractures is expected to dominate transport behavior, primarily because the expected flow rates through these systems exceed the matrix flow capacity under a unit gravitational gradient. Advection through fractures is fast because of high permeability and low porosity, with few opportunities for radionuclides to contact rock matrix. A few of the hydrogeologic units have much larger matrix permeability and are expected to capture most of the fracture flow by advection from the fractures to the matrix,

causing much slower transport velocities and closer contact of the radionuclides with the matrix. Advective transport pathways result from and therefore follow the flow pathways, which are predominately downward. However, lateral diversion is expected along hydrogeologic unit contacts having strong contrasts in rock properties, particularly in areas of perched water. Flow that is diverted laterally ultimately finds a pathway to the water table through more permeable zones, which may be faults.

The detailed geometry of fractures and matrix pore spaces at Yucca Mountain is far too complex to be modeled explicitly. On the other hand, it is important to capture the larger-scale spatial variability, such as differences between welded and nonwelded hydrogeologic units, and the differences in fracture and matrix properties at the local scale. To show this variability, a dual-permeability model is used for fractured rock. In the dual-permeability model, the fractures and matrix are distinct interacting continua that coexist at every point in the modeling domain. Each continuum is assigned its own hydrologic properties such as permeability and porosity, which may also vary spatially. In general, the fractures are modeled as a highly permeable continuum having low porosity; the matrix is modeled as a much less permeable continuum having higher porosity. The dual-permeability model offers a bimodal approximation to the true spectrum of fracture and matrix properties. More importantly, the dual-permeability model can capture the effects of fast pathways for radionuclide transport from the repository to the water table. This feature is an important improvement over single-continuum models (Robinson et al. 1996, Section 9.5.3; Robinson et al. 1995, Section 8.3).

The conceptual model for UZ transport is strongly tied to the conceptual model for UZ flow. As described above, advective transport because of flow is the main transport mechanism that can move radionuclides from the repository to the water table. Conceptual models for UZ flow are commonly based on a continuum relationship, known as Darcy's law, which relates volumetric flow rate and the gradient in hydraulic potential. In the case of fractured, porous rock such as the volcanic rock that constitutes Yucca Mountain, these continuum relationships are extended to embrace two coexisting continua, fractures and rock matrix, that interact according to the same constitutive relationships that govern flow in a single continuum. From the standpoint of UZ transport, the need for explicit and separate representation of fracture and matrix flow is because of the extreme disparity in transport velocities that can occur in the two continua. Travel times for radionuclides transported to the water table exclusively in fractures are expected to be about 10,000 times faster than travel times for radionuclides moving exclusively in the matrix. In addition, the transport velocities in the fractures may be sufficiently rapid that radionuclide concentrations in the fractures are in disequilibrium with the matrix. For example, a high concentration of radionuclides entering fractures at the repository may penetrate the entire UZ before establishing a uniform, equilibrated concentration in the rock matrix. Therefore, an explicit and dynamic model of transport through the fractures and matrix, as well as exchange between the fractures and matrix, is needed to represent the system. The dual-permeability model provides the necessary level of detail to capture the important differences between transport through fractures and matrix. There are other possible approaches to modeling flow in fractures and matrix rock. However, these other models either don't recognize the important dynamic coupling between fractures and matrix, such as the ECM, or are impractical to implement at the field scale, such as the discrete fracture model (particularly for systems with advective transport in the fractures and matrix, such as a dual permeability system). For these

reasons, the flow and transport models for the UZ are based on dual permeability. (Robinson et al. 1997, Section 4.7.1).

In general, flow in the UZ is time dependent or transient. One mechanism responsible for this time dependence is the time variations in the infiltration flux at the surface. The time variation of the infiltration flux may be approximated as occurring over short intervals characterized by changes in weather, resulting in episodic transient flows, or over much longer time periods corresponding to climate changes. Existing information concerning episodic transient flow seems to indicate that it may not be able to frequently penetrate through the UZ to the level of the repository because of the dampening influence of the Paintbrush nonwelded (PTn) hydrogeologic unit. Episodic transient flow propagating through fractures tend to be absorbed by the PTn unit, resulting in much slower drainage of the episodic flows in lower hydrogeologic units at and below the repository level (Robinson et al. 1997, Section 6.13). The total quantity of water able to penetrate the UZ in the fracture system without interacting with matrix may, in fact, be larger for steady flow than for transient pulses, given the same average infiltration flux (CRWMS M&O 1998, Section 7.6). For these reasons, episodic transient flows have not been incorporated into the TSPA-VA calculations.

Changes in unsaturated flow because of longer-term changes in climate have a more pronounced influence on UZ flow than episodic transient flow. Sustained changes in infiltration associated with climate change ultimately impact the entire flow field in the UZ. The actual transient period during which the UZ flow responds to a climate change, however, has been found to be less significant (Robinson et al. 1997, Section 8.11; CRWMS M&O 1998, Section 7.4). The reason is that the change in flow in the fractures, which dominates the flux in most hydrogeologic units, responds relatively quickly to a change in infiltration. Therefore, the quasi-steady flow model was used to estimate the effects of climate change on radionuclide transport. In this model, infiltration rate is assumed to change abruptly when climate changes from one steady flow field to another. Transport calculations simply re-start when climate changes. A distributed source of radionuclides throughout the UZ is derived from transport calculations using the flow field for the previous climate, and a new steady flow field is selected based on the new climate.

In addition to the change in the UZ flow field, the location of the water table is also assumed to change abruptly at the time of climate change. The three climate states—present day, LTA, and super pluvial (see Section 3.1)—have successively higher water table elevations in response to the increasing infiltration. If the water table rises with the climate change, the radionuclides in the UZ between the previous and new water table elevations are immediately available for SZ transport. In the TSPA-VA model, water table elevations change by 80 m from present-day to long-term-average climates and by 120 m from present-day to super-pluvial climates.

The dominant flow direction is expected to be mainly downward over large scales. However, local flow field variations are expected to be 3-D. The importance of these variations lies primarily in the kinds of rock units and fracture characteristics that dominate along the 3-D flow paths. A secondary consideration is that 3-D flow paths from the repository to the water table will necessarily be longer than strictly vertical flow paths. Three-dimensional flow patterns are expected along rock unit contacts with contrasting properties, particularly in zones where these contrasts are believed to be features that create perched water (see Figure 7-1). To capture these

effects, the flow and transport calculations are performed in three dimensions. Spatial variability is captured in the 3-D relationships of the hydrogeologic units and structural features, for example, faults, and in the variations in hydrogeologic and transport properties assigned to the hydrogeologic units.

7.2.2 Matrix Diffusion

Diffusion is the movement of dissolved or colloidal material because of random molecular motion. It is not an effective mechanism for transport between the repository and the water table because of the large distance involved (about 300 m). However, diffusion can play an important role in radionuclide exchange between fractures and rock matrix. In this case, molecular diffusion affects the persistence of a dissolved ion in the fracture flow stream. The relative influence of this mechanism on overall transport through the UZ depends on the rate of movement through fractures as well as the degree of fracture/matrix contact.

Bulk diffusive flux occurs when concentration gradients are present because diffusion is driven by random molecular motion. In addition, the matrix diffusion coefficient is a function of the free water diffusion coefficient, temperature, radionuclide mass, atomic or molecular dimension, and charge, as well as the matrix pore structure and water saturation. The temperature variations are expected to be small over the time period for radionuclide releases to the UZ. The effects of pore structure and water saturation have been shown to depend primarily on the volumetric water content of the rock (LANL 1997b). For rock in the UZ, the water content is relatively uniform spatially. Therefore, as a simplification, variations in the matrix diffusion coefficient are assumed to be primarily dependent upon the radionuclide type, that is, mass, size, and charge. In this case, measurements indicate that the primary difference is between cationic and anionic radionuclides (Triay et al. 1997, Section VI). Anionic radionuclides have lower matrix diffusion coefficients than cationic radionuclides; that is, they are transported more slowly by diffusion. Lower coefficients for anionic radionuclides are believed to be a result of size and charge exclusion of the anionic radionuclides from a portion of the pore structure. The surfaces of the pore minerals are generally negatively charged under the chemical conditions of the undisturbed environment.

In general, the direction of radionuclide transport by matrix diffusion between the fracture continuum and the matrix continuum depends on the direction of the concentration gradient. However, the most important influence of matrix diffusion is expected to be on radionuclide transport through fractures. Radionuclides traveling through fractures are always near the matrix relative to the fracture spacing. This proximity is a result of the fracture and matrix geometry, where the ratio of fracture aperture to fracture spacing is small. Also, diffusive penetration of the matrix is proportional to the square root of the fracture transport time. Because of relatively fast transport through fractures, most radionuclides diffusing from fractures into the matrix do not penetrate far, relative to the fracture spacing. For this reason, the effects of fracture spacing on matrix diffusion are expected to be negligible, and the effects of finite fracture spacing may be ignored. Similarly, because of the relatively small fraction of matrix affected by matrix diffusion (that is, diffusive movement from fractures into the matrix is relatively slow) an approximate representation can be made for fracture transport, including matrix diffusion, separately from the advection of radionuclides through the matrix. Detailed development of the fracture transport model including matrix diffusion is given in Section 7.4.

7.2.3 Dispersion

Dispersion is a transport mechanism caused by localized variations in flow velocity. These variations cause dispersion of the radionuclides both along and transverse to the average flow direction. Variations in both the magnitude and direction of the velocity contribute to the overall dispersion. This dispersion, under certain limiting conditions, can act in a manner analogous to diffusion, in which mass flux is proportional to the concentration gradient. Dispersion is most important where concentration gradients are the largest—near the front of a propagating plume or along the lateral edges of the concentration field. Dispersion smears sharp concentration gradients and can reduce the breakthrough time, or arrival time at a specific point, for low concentration levels of an advancing concentration front.

Dispersion is included in the transport conceptual model using a standard relationship based on Fick's law between mass flux and concentration gradient (Robinson et al. 1997 Section 4.4.1). The dispersion coefficient in the Fickian relationship is expressed as the product of the mixing length scale, or dispersivity, and the average linear velocity. Dispersion is independently represented in both the fracture and matrix continua. However, dispersion is not expected to play an important role in the UZ transport. The repository emplacement area is very broad, relative to the distance to the water table, and this geometrical arrangement tends to suppress dispersion effects. Longitudinal dispersion becomes secondary to the explicitly modeled variations in transport velocities across the potential repository because of variations in infiltration. Lateral dispersion is limited by the short transport path from the repository to the water table compared with the size of the source. Also, the explicitly modeled variations in transport velocity caused by the fracture/matrix system also tend to dominate dispersion.

7.2.4 Radionuclide Sorption

Sorption is the general term for describing a combination of chemical interactions between the dissolved radionuclides and the solid phases (that is, either the immobile rock matrix or colloids). Any given sorptive interaction is caused by a set of specific chemical interactions such as surface adsorption, precipitation, and ion exchange. However, the sorption approach does not require identifying the specific underlying interactions. Instead, batch sorption experiments are used to identify the overall partitioning between the aqueous and solid phase, characterized as a "sorption" or distribution coefficient (K_d). The strength of the sorptive behavior is a function of the chemical element, the rock type involved in the interaction, and the geochemical conditions of the water contacting the rock. Sorption reduces the rate of advance of a concentration front in advective and diffusive transport, and amplifies the diffusive flux of radionuclides from the fractures to the rock matrix through its influence on the concentration gradient.

Numerous rock-water chemical interactions may influence radionuclide transport (Triay et al. 1997, Section C):

- *Ion adsorption*—metal cations sticking to mineral surfaces due to London-van der Waals forces and hydrogen bonding

- *Ion exchange*—substitution of an aqueous cation for the cation in a mineral structure

- *Surface complexation*—coordination of an aqueous cation with a deprotonized metal hydroxide at the mineral surface

- *Precipitation*—generation of a bulk solid phase.

The nature and strength of these rock-water interactions are highly dependent on the chemical composition of both the aqueous and solid phases. The conceptual model used to capture all these interactions and sensitivities is the minimum K_d model. This model bounds the distribution of radionuclides between the mobile, or dissolved in the aqueous phase, and immobile, attached to the solid phase, radionuclides using a linear, infinite-capacity partitioning model. In this model, the sorbed, or immobile, concentration is equal to the aqueous concentration times the partitioning coefficient (K_d). Because of the numerous mechanisms and dependencies known to influence sorption, using a linear partitioning model with a single coefficient provides only a minimum bound for K_d, and hence the immobilized fraction.[1]

The range of pore-water compositions of the matrix of the undisturbed (that is, pre-repository) UZ is expected to overlap the combined range of groundwater compositions for the tuffaceous aquifer and the deeper carbonate aquifer (Meijer 1992, pp. 18-19). Therefore, sorption behavior investigated under this range of conditions is used to bound sorption in the UZ (Triay et al. 1997, p. 173). Under the assumptions of the model, the minimum K_d approach is conservative for radionuclide transport, except in the case of colloid-facilitated transport, which is discussed in Section 7.5.3.6.

The surfaces of fractures, often lined with minerals that differ from the bulk of the rock matrix, may be capable of sorbing many of the radionuclides (Triay et al. 1997, p. 173). However, there has been limited characterization of the distributions of the fracture-lining minerals and sorptive interactions with these minerals. Also, the fracture minerals have a relatively small volume and surface area. For these reasons, it is conservatively assumed that there is no sorptive interaction with the fracture surfaces, and sorptive interactions are only possible for radionuclides in the matrix continuum.

Sorption is known to be sensitive to rock mineralogy and texture because these characteristics affect the types and available areas of different mineral surfaces. Although many different mineral types are present in Yucca Mountain, three basic rock types have been identified as having distinct sorptive interactions with the different elements of the waste form. These rock types are devitrified tuff, vitric tuff, and zeolitic tuff. Sorption coefficients for all radionuclides are specified for each of these rock types in the TSPA-VA conceptual model.

Sorption coefficients for performance assessment have been chosen based on site-specific batch sorption data, sorption data available from the general literature, and expectations based on the expected mechanisms for sorption (Triay et al. 1997). This information has been used to establish average minimum bounds for radionuclide sorption under a variety of expected

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[1] "Minimum bound" means that the K_d used in the model represents the smallest reasonable ratio of radionuclides attached to the solid phase versus the aqueous phase. This assumption is conservative because it implies maximum transport of radionuclides in the aqueous phase.

geochemical conditions (for the natural system) on the identified rock types. The range of distribution coefficients selected are, therefore, expected to be conservative in terms of the resulting predictions of sorption for radionuclide transport.

Most of the investigated radionuclides are sorbed most strongly by the zeolitic tuff. Zeolitic tuff is primarily composed of clinoptilolite, and usually includes opal-CT (cristobalite - tridymite), quartz, and feldspar (Triay et al. 1997, Section I.D). Vitric tuffs, primarily composed of glass and feldspar, are generally less sorptive than zeolitic or devitrified tuffs. Devitrified tuff, also known as welded tuff, is primarily composed of alkali feldspar and tridymite. However, the importance of sorption in the different rock types is not only a function of the sorptive strength, K_d, but also the degree of exposure the radionuclides have with the rock matrix during transport through the UZ. The relative amount of radionuclide transport through the matrix is a strong function of the matrix permeability, where radionuclide transport is favored through a matrix with higher permeability. The vitric tuff is typically much more permeable than the devitrified or zeolitic tuffs, providing greater radionuclide contact with the vitric matrix.

Variations of geochemical conditions in time and space will affect the characteristics of radionuclide-rock interactions. Geochemical variability may be separated into two broad categories. One type of variability may be attributed to natural processes of the "undisturbed" environment that are observed in water samples from different locations. The method for determining the appropriate minimum K_d is based on the range of geochemical conditions representative of this natural variability. Sensitivity studies suggest that future natural variations in geochemical conditions because of climate change will have little influence on radionuclide transport (Robinson et al. 1997, pp. 11-48). A second type of variability is caused by the thermal-chemical interactions of the repository with the geochemical/mineralogical environment, disturbed environmental conditions. Although sensitivity studies indicate that transient thermal effects have little influence on radionuclide transport (Robinson et al. 1997, pp. 11-48 and 11-49), potential longer-term effects on geochemical and mineralogical conditions require further investigation (Section 7.7).

7.2.5 Colloid-Facilitated Transport

Colloids are potentially mobile in water flowing through the UZ. Colloids, because they are small solids, can interact with radionuclides through sorption mechanisms. Unlike sorption of radionuclides to the rock matrix, however, radionuclides sorbed on colloids are potentially mobile. Therefore, colloidally-sorbed radionuclides can transport through the UZ at a faster rate than the aqueous species without colloids. Another form of colloidal radionuclide movement occurs when the radionuclide is an integral component of the colloid structure (see Section 3.5). In this case, the radionuclide is irreversibly bound to the colloid, as compared to the typically reversible sorption mechanism. The different types of colloids considered in the performance assessment model are clay colloids, iron oxy-hydroxy colloids, spent fuel waste form colloids, and glass waste form colloids. Specifics concerning these different types of colloids and their interactions with radionuclides are given in Chapter 6, Section 6.4.2.

The transport equation of any aqueous radionuclide through fractures is expressed in terms of the previously mentioned processes of advection, sorption on fracture surfaces, if included, matrix diffusion, dispersion, and radioactive decay. If the radionuclide is sorbed on colloids, then

additional terms are needed in the equation to describe the advection, sorption, dispersion, and decay of the radionuclides on colloids. Colloid particles themselves are not expected to experience any significant matrix diffusion because of the very low diffusion coefficients associated with colloids. It is assumed that sorption of radionuclides on colloids may be approximated using the K_d conceptual model described above for sorption, except that the conservative bound now is the maximum K_d. A maximum K_d for sorption on the colloidal minerals is a conservative bound because more of the radionuclides are placed on the colloids. Maximum K_ds were identified using desorption experiments. These will give high values for K_d because the kinetics of desorption are slow, and will therefore tend to lead to an excess of radionuclide that is considered to be sorbed at equilibrium. Advective radionuclide transport through the fractures is enhanced by reduced matrix diffusion and matrix sorption. Given a steady, uniform concentration of colloids and the K_d model for sorption, the ratio of the radionuclide mass in aqueous and colloidal form is a constant value (K_c). Both the linear nature of the transport equations for aqueous and colloidal forms and the linear partitioning of the aqueous and colloidal concentrations allow simplification of the transport equation. The mathematical transport model for aqueous and colloidal radionuclides may be reduced to a single equation in terms of the aqueous radionuclide concentration (Robinson et al. 1997, Section 8.10). Additionally, no new terms appear in this transport equation, as compared with strict aqueous transport. Therefore, the new transport equation for reversibly sorbed aqueous-colloidal transport has the same structure as that for strict aqueous transport. The difference is that the transport coefficients now include information concerning the colloidal partitioning.

Colloids may not be able to move through the some of the rock matrix, particularly the welded and zeolitic rock types, because of the colloid size relative to the matrix pore size. However, movement through the more permeable, nonwelded vitric rock in the Calico Hills may be possible. Advective, aqueous-phase transport of radionuclides between fractures and matrix is only a strong feature of the flow field in the higher-permeability Calico Hills vitric unit. As an approximation to simplify the transport model, colloids are also allowed to move by advection with the aqueous-phase radionuclides between the fractures and matrix. However, colloids are not allowed to exchange between fractures and matrix through matrix diffusion (Section 7.4.5.1).

In some cases, radionuclides may be an integral part of the colloid structure itself, causing stability of the radionuclide attachment to the colloid that may be approximated by an irreversible sorption model. To model the transport of these irreversibly sorbed radionuclides, the transport model described above for reversibly sorbed, colloid-facilitated transport is modified to account only for transport processes that affect colloids. In other words, for this portion of the radionuclide mass, interaction with the aqueous phase and immobile rock phase is neglected. With this simplification, colloid transport is mathematically analogous to aqueous radionuclide transport with no matrix diffusion and no sorption (Section 7.4.5.2).

7.2.6 Radioactive Decay

Radioactive decay is a process that affects the concentration of radionuclides during transport through the UZ. For simple decay, the radionuclide concentration decreases exponentially with time, creating decay products that are stable. Chain-decay adds another layer of complexity because of the ingrowth of new radionuclides created from the decay of a parent radionuclide.

Radioactive decay is modeled using simple exponential decay. For the TSPA-VA base case, ^{237}Np, ^{234}U, and ^{231}Pa are the only radionuclides significantly affected more than a few percent by the neglect of the chain-decay processes. Consideration of chain decay causes transport calculations to be more complex because daughter radionuclides are created during transport of the parent isotope. The creation of progeny could be modeled in a complex calculation as a distributed source term throughout the modeling domain. However, a simplified approach developed specifically for TSPA calculations is used that adjusts the initial radionuclide source term (the inventory in the repository at the start of the TSPA-VA calculations) to compensate for the production of the radionuclides ^{237}Np, ^{234}U, and ^{231}Pa. In the case of ^{237}Np and ^{234}U, where the parent radionuclides of concern (for ^{237}Np these are ^{241}Am, ^{241}Pu, and ^{245}Cm, while for ^{234}U the parents are ^{238}Pu, ^{242}Cm, and with a mass factor of 0.82 for a branching chain ^{242}Am) have a shorter half-life, it was assumed that the parents had decayed to the isotopes of interest at the start of the simulation. This was achieved by defining the mass of each of the above defined parents present in the waste forms per MTU and transforming these mass concentrations (by the ratio of isotopic mass) to the effective concentration of the radionuclide considered. However, for ^{231}Pa the half-life is much shorter than that of the parent, ^{235}U. To allow for the neglect of decay, it was assumed that the protactinium was, at all time, in secular equilibrium with the uranium parent at the source, that is, for every curie of uranium in the waste form there was a curie of protactinium. In the RIP code, this was done by providing the code with the mass (not activity) per waste package of ^{231}Pa that would be in secular equilibrium with the ^{235}U. To avoid an unnatural "rapid" decay of the ^{231}Pa, the half-life of ^{235}U was used to characterize the decay of the ^{231}Pa.

7.2.7 Thermal-Hydrologic, Thermochemical, and Thermomechanical Processes

The potential repository will perturb the natural system due to the introduction of waste heat and foreign materials. Thermal-hydrologic modeling is done for TSPA-VA to better define the temperature, humidity, and water contact conditions that will affect waste package corrosion, waste form dissolution, and radionuclide releases from the engineered barrier system. The thermal-hydrologic conditions also affect the flow field, and hence the radionuclide transport, through the UZ. However, the thermal-hydrologic process is most significant over the first 2,000 to 3,000 years following waste emplacement. Sensitivity studies indicate (see Section 7.6) that most of the radionuclide transport through the UZ will take place after thermal-hydrologic activity has subsided. Thermal-hydrologic processes are not expected to substantially alter UZ radionuclide transport as compared to transport through the undisturbed system. Therefore, the effects of thermal-hydrologic processes are not included in the UZ radionuclide transport model.

Introduced materials and thermochemical interactions may result in persistent changes to the geochemical and mineralogic environment in the UZ. These effects could, for example, alter UZ sorption characteristics. Thermomechanical and thermogeochemical interactions could also significantly affect hydrologic properties through fracturing under thermal expansion and plugging of flow paths due to precipitation/dissolution reactions. Thermal expansion and fracturing of the PTn may alter the characteristic of flow deep into the UZ. Thermal alteration of the Topopah Spring basal vitrophyre may result in a less permeable condition due to glass alteration. Modeling of these more complex coupled processes is very limited at this time. The current approach to bound these effects is to investigate, as sensitivity studies for TSPA-VA, a wide range of possible flow conditions (e.g., fracture/matrix contact) and consider the effects of

thermochemical perturbations on sorption using bounding assumptions (e.g., no sorption of actinides due to high pH).

7.3 ISSUES ASSOCIATED WITH PROCESS

7.3.1 Development of Issues

A workshop concerning the treatment of UZ radionuclide transport for Total System Performance Assessment - Viability Assessment (TSPA-VA) was held in Albuquerque, N.M. on February 5-7, 1997. The workshop addressed the attributes of radionuclide transport in the UZ that are important to the long-term performance of a geologic repository at Yucca Mountain (CRWMS M&O 1997).

The development of model abstractions requires collaboration between TSPA and process-level modelers as well as those who do laboratory and field measurements. This collaboration is required to ensure that the abstractions developed are as consistent as possible with the most comprehensive and up-to-date understanding of relevant site characteristics and processes. Given this multi-dimensional viewpoint represented by the different participants, the objectives of the UZ radionuclide transport abstraction/testing workshop were to develop the following:

- A comprehensive list of UZ radionuclide transport issues that are important to long-term performance. (The purpose of this objective is to identify processes, process models, observations, and parameter ranges that are pertinent to UZ radionuclide transport and may be important to long-term repository performance.)

- A ranking of the importance of these issues using the collective expertise of the assembled participants. (The ranking was performed relative to criteria that reflect impact on long-term performance. Ranking will provide (1) provide guidance on which issues to focus on during the remainder of the workshop and (2) documentation concerning the perceived importance of the issues identified.)

- Draft work proposals to address the key issues identified that are not adequately constrained by current TSPA models. (In general, the work proposals will answer some question about UZ radionuclide transport processes that could be used to justify or develop a process abstraction or used to define the range of different parameters that are needed to bound system performance.)

Most of the workshop attendees have participated in previous performance assessment or site characterization studies that address or are linked to UZ transport. The workshop was structured to discuss, extract, and prioritize the important facets of the problem from the collective viewpoint of these workshop attendees. Therefore, brief presentations of previous, current, and proposed work were given by the workshop attendees to stimulate discussion and identify the important system features that require further study.

The workshop was divided by subject matter into the categories physical transport processes; chemical interactions and repository-perturbed environment; and heterogeneity. A list of primary issues for each category were identified (CRWMS M&O 1997).

Criteria were developed so that the issues could be ranked against their potential effects on long-term performance. The primary effect of UZ transport on long-term performance is through the spatial and temporal distributions of radionuclide travel time to the water table and radionuclide mass flux at the water table. Travel time is potentially important for meeting requirements linked to specific time frames, whereas mass flux is a principle factor affecting dose. Although travel time and flux are linked through the water flux, these factors can vary independently. The other components of travel time and mass flux are radionuclide velocity and concentration. Therefore the four criteria chosen for ranking the issues were radionuclide concentration, radionuclide velocity, water flux, and the distribution of travel times to the water table.

7.3.2 Issues and Ranking; Physical Transport Processes

The highest-ranking issues in the area of physical transport processes were found to be (1) fracture/matrix interaction and (2) the treatment of long-term transient flow associated with climate change. The relative degree of transport in fracture versus matrix has a large impact on the movement of fronts through the UZ, hence on the delay of radionuclides during transport. Several features of unsaturated flow and transport have a strong influence on the partitioning of transport between the fractures and matrix, and many of the model parameters that influence the quantitative behavior are uncertain. Therefore, the effects of these parameters on subsystem behavior were identified for sensitivity studies to determine the ranges and combinations of parameters that are important in the unsaturated-zone transport process. The results of these sensitivity studies are given in Section 7.6.1.

Climate change and its effects on UZ flow and transport were recognized by the workshop attendees. The principle issue was the development of an appropriate way to integrate the effects of long-term changes in UZ flow on transport for a model that was based on a steady flow approximation. The solution to this problem was to use a quasi-steady flow approximation in the transport model. The quasi-steady flow approximation is that flow fields switch instantaneously from one steady flow condition to another, based on the changes in infiltration rate with changes in climate. The transient flow periods associated with changing from one infiltration rate to another are ignored. Details concerning how this approximation is used in the transport model, along with a comparison with the fully transient solution, is given in Section 7.4.4.

Longitudinal dispersion was not believed to be as important by the workshop attendees, given that the transport model would use a dual-permeability formulation in which the fracture and matrix contributions to the transport are modeled explicitly. Mixing of the radionuclide releases laterally, upon reaching the water table, were assumed to be reasonably complete. However, the radionuclide mass flux at the water table was believed to be relatively independent of lateral mixing, and the primary performance attributes of the UZ were assumed to be radionuclide mass flux and travel times at the water table. Analyses concerning the sensitivity to longitudinal dispersion are given in Section 7.6.1.2.6.

7.3.3 Issues and Ranking: Chemical Interactions and Repository-Perturbed Environment

The highest ranking issues concerning chemical interactions and the repository-perturbed environment were found to be (1) justification of the linear sorption method relative to more

complex chemical interactions, (2) effects of colloid-facilitated radionuclide transport, and (3) thermal-chemical mineral alteration due to the repository-perturbed environment.

Sorption plays an important role in delaying the arrival of many radionuclides that may potentially be released from the engineered barrier system. The sorption model used for TSPA UZ radionuclide transport is a linear, infinite capacity sorption model. Chemical interactions characterized as sorption in batch sorption experiments are known to result from a variety of different chemical mechanisms such as ion adsorption, ion exchange, surface complexation and precipitation/dissolution. An investigation concerning the range conditions under which this sorption model is appropriate is presented in Section 7.6.3.

Colloid-facilitated radionuclide transport may greatly reduce the time for radionuclide migration through the UZ by reducing or eliminating sorptive interactions with the rock matrix and by concentrating radionuclide transport in the fracture continuum. The model used for radionuclide transport through the UZ, including the effects of radionuclide attachment on colloids, is described in Section 7.4.5.

Thermal-chemical alteration of minerals and of geochemical conditions are potentially important to radionuclide transport through the UZ. Some initial studies on the effects of thermal-chemical changes due to interactions between repository heat and materials with the UZ environment are presented in Section 7.6.3.

7.3.4 Issues and Ranking: Heterogeneity

The highest ranking issues concerning heterogeneity were found to be (1) the influence of lateral diversion of radionuclide pathways, (2) greater spatial resolution of stratigraphic characterization for UZ flow and transport, and (3) fine-scale heterogeneity of zeolitic alteration.

Issues concerning lateral diversion have been directly addressed in TSPA-VA through the 3-D UZ flow and transport model. Some of the results of lateral diversion in the 3-D UZ transport model are discussed in Sections 7.5.2 and 7.6.1.2.7.

Issues regarding the finer-scale structure of the stratigraphy and zeolitic are concerned with the potential for radionuclides that may strongly sorb to some rock types, particularly zeolites, to by-pass these rock types due to finer-scale heterogeneity that is not explicitly represented in the flow and transport models. A study concerning the effects of fine-scale heterogeneity in zeolitic alteration on radionuclide transport is presented in Section 7.6.5.

7.4 APPROACH AND IMPLEMENTATION OF ABSTRACTION FOR PROCESS

The approach used for TSPA-VA is given in detail in this section. Details of the particle tracking method and its coupling with the total system performance assessment code, RIP, are discussed, along with the parameters needed for base-case TSPA calculations. This section also provides detailed discussions of process abstractions to be used with the particle-tracking model. The process abstractions address the treatment of long-term transient flow due to climate change and colloid-facilitated radionuclide transport. Much of this section is also found in Robinson et al. (1997), Chapter 4.

7.4.1 Unsaturated Zone Transport Conceptual Model And Mathematical Approach

The conceptual model used for treating the interaction of flow and transport through fractures and matrix is important for both the distribution of flow between fracture and matrix and for the solute transport velocities that are computed. The simplest model of fracture/matrix interaction is the equivalent continuum approximation. In this model, the capillary pressures are assumed to be equilibrated such that the saturation (and hence flow) distributions may be determined from a single independent variable (e.g., the fracture saturation or the matrix saturation). The equilibration of capillary pressure over the two continua represents an asymptotic endpoint for steady flow in a homogeneous, semi-infinite fracture/matrix system, where the flow is initiated at some boundary and partitioned between the fracture and matrix continua in some ratio that is not in equilibrium.

For flow that is initiated (at the surface) in the fractures, the equivalent continuum approximation will underestimate the amount of fluid passing through the fractures. The degree of error is dependent on the relative rates of flow through the fractures and imbibition from the fractures into the matrix.

The equilibration concept may also be applied to the distribution of solute between flow in the fractures and matrix. In this case, the solute equilibration depends on an additional mechanism, molecular diffusion of the solute within the rock matrix. This additional mechanism is important for solute transport because the equilibration of capillary pressure between fractures and matrix does not ensure that solute concentrations are likewise equilibrated. This is primarily due to the fact that water imbibed from fractures into matrix will tend to equilibrium according to the propagation of water saturation variations under the influence of capillary pressure. The aqueous solution associated with this capillary imbibition process will (miscibly) displace the existing matrix water. Molecular diffusion is then needed to equilibrate solute concentrations over the matrix. As it turns out, the equilibration of capillary pressure is typically faster than the redistribution of solutes due to matrix diffusion. Therefore, during imbibition solute concentration fronts in the matrix will separate from and lag behind water saturation fronts. This also means that even if flow may be reasonably represented by an ECM, transport may still require a dual permeability approach.

For flow and dissolved solute initiated at a boundary in the fractures, an equivalent continuum approximation will likewise underestimate the amount of solute flux passing through the fractures. Due to the high permeabilities and low porosities associated with the fracture system as compared with the matrix system, the ECM will also underestimate the average solute velocities.

These considerations lead to the use of a dual permeability model, which accounts for the rate of exchange between the fracture and matrix continua based on the appropriate driving forces of capillary pressure and solute concentration gradients.

7.4.1.1 Mathematical Models Based on Differential Material Balance Equations

The traditional method of computing the flow and transport of water and dissolved solutes in geologic media, including fractured rock, is through the solution of differential equations that

govern the flow of water and transport of solutes. The equations are based on differential mass conservation concepts combined with differential relationships between the state of a system and the flux of water or solute that is expected (e.g., Darcy's law or Fick's law).

The resulting differential equations governing solute transport in a fracture/matrix system are advection-dispersion equations for each continuum, coupled through solute exchange due to advective and diffusive mechanisms. Traditional numerical solutions to the advective-dispersion equation, such as finite-element or finite-difference codes, are versatile and allow the simultaneous solution for multiple interacting species. One drawback of a finite-difference or finite-element solution to the advection-dispersion equation is that significant numerical dispersion may arise in the portion of a computational domain occupied by a front of rapidly varying concentration. Reducing the numerical dispersion requires either increased grid resolution or higher-order approximation methods, both of which may lead to prohibitive computational costs. Numerical dispersion is identical in character to actual dispersion, so it is difficult to separate numerical from actual dispersion in the results of complex transport simulations.

Approaches to cope with this problem include front-tracking algorithms with multiple grids (e.g., Yeh 1990; Wolfsberg and Freyberg 1994), the method of characteristics (e.g., Chiang et al. 1989), hybrid Eulerian-Lagrangian solution techniques (Neuman 1984), and particle tracking techniques (e.g., Tompson and Gelhar 1990). Front-tracking algorithms solve the advection-dispersion equation in integrated form on a numerical grid while tailoring the mesh to increase the resolution of the calculation at fronts. In contrast, an Eulerian-Lagrangian technique (Neuman 1984) casts the advection-dispersion equation using the total derivative, so that the advection portion of transport can be solved accurately using particle-tracking techniques or the method of characteristics, while the dispersion component of transport is solved on a finite-difference or finite-element grid using standard techniques. A particle tracking method, as discussed in the next section, has been chosen for TSPA-VA UZ radionuclide transport calculations.

7.4.1.2 Mathematical Models Based on Particle Tracking; Pathline Models and Residence Time/Transfer Function Models

Particle-tracking transport models take a fundamentally different approach from the differential material balance method discussed above. The trajectory of individual molecules or packets of fluid containing molecules are tracked through the model domain. When the fluid path lines are the model result of interest (Pollack 1988; Lu 1994), a relatively small number of particles can be used to trace the streamlines. Particle tracking is also used to simulate solute transport, such as the migration of a contaminant plume (Akindunni et al. 1995) or the prediction of breakthrough curves in interwell tracer experiments (Johnson et al. 1994). For these applications, a relatively large number of such particles must be used to obtain accurate solutions to the transport problem. Particle tracking has also been used to solve the advective portion of complex reactive transport models that simulate chemical reactions among multiple species (Fabriol et al. 1993).

In a typical particle tracking algorithm, a particle is sent to a new position assuming that the magnitude and direction of the velocity vector are constant during a time step dt. If small

enough time steps are taken, particle pathways can be tracked accurately. Dispersion is treated as a random process that diverts the particle a random distance from its dispersion-free, deterministic path. In these so-called "random walk" models (e.g., Kinzelbach 1988), dispersion is usually calculated stochastically subject to a Gaussian model to reproduce the specified dispersion coefficient. The technique has also been extended by employing non-Gaussian random walk functions to represent scale-dependent dispersion (Scheibe and Cole 1994). Linear, equilibrium sorption can be handled through the use of a retardation factor to correct the magnitude of the particle velocity.

A crucial component of all random-walk particle-tracking algorithms developed to date is the need to accurately estimate the velocity at every position in the model domain. In the context of a finite-difference or finite-element numerical code, this means that velocities at positions other than the node points of the fluid flow grid must be computed using an interpolation scheme. Many studies have proposed and studied the accuracy of different interpolation schemes, including methods developed for regular, two- or 3-D finite difference grids (Goode 1990; Schafer-Perini and Wilson 1991; Zheng 1993), two- and 3-D finite-element grids (Cordes and Kinzelbach 1992), and for codes that employ the boundary element method for computing fluid flow (Latinopoulos and Katsifarakis 1991). Special techniques have been developed to handle complexities such as point fluid sources and sinks and abrupt changes in the conductivity of the medium (Zheng 1994).

In the process of adding complexity to the particle-tracking algorithms, some of the intuitiveness of the original particle-tracking concept has been lost. Furthermore, many of the velocity interpolation schemes are computationally intensive, thus limiting the number of particles that can practically be used. Another drawback to traditional particle-tracking approaches is that spatial and temporal discretization often results in numerical inaccuracy in the fluid flow solution upon which velocity determinations are based. Thus, precise and time-consuming velocity interpolation schemes may not be justified in finite-difference or finite-element models. Finally, for dual porosity models that employ overlapping continua to represent fracture and matrix flow (Zyvoloski et al. 1992; Zimmerman et al. 1993), velocity interpolations on each continuum must be coupled to a transfer term that allows particles to move from one medium to the other. This additional complexity, along with the approximation associated with the dual-porosity method itself, may make precise velocity interpolation calculations of limited validity.

In this chapter, a new particle-tracking technique is developed for transient, multi-dimensional finite-difference or finite-element codes. The algorithm is designed for computing solute concentration fields quickly and easily with structured or unstructured numerical grids of arbitrary complexity. Both continuum and dual-porosity formulations can be simulated. This flexibility is accomplished by adopting a new strategy for mapping out the path of a particle, one that replaces the calculation of an "exact" pathline with a cell-to-cell migration of the particle. The mass flux from cell to cell is used directly, and no velocity interpolations are required. Since numerical solutions for fluid flow are typically mass-conservative (though not necessarily accurate), the particle-tracking method automatically conserves mass. Despite the advantages of the new particle tracker discussed here, there are still some inherent numerical limitations of the method. These are primarily that large numbers of particles are commonly required to reduce the noise in the computed responses and that empirical tests are needed to determine convergence of the calculations.

7.4.2 Implementation of Particle Tracking in FEHM

The particle-tracking method developed in the present study views the fluid flow computational domain as an interconnected network of fluid storage volumes. Particles travel only from cell to cell, requiring no greater resolution of the particle pathways. In this sense, the method is similar to the node-to-node routing method of Desbarats (1990). This simple starting point has been extended to include many different transport submodels and complex flow domains. The description that follows is applicable for steady-state, single-porosity flow fields; the corrections to the method for treating transient flow systems and dual-porosity model formulations are discussed in subsequent sections. The two steps in the particle-tracking approach are: (1) determine the time a particle spends in a given cell and (2) determine which cell the particle travels to next. These two steps are detailed below.

The residence time for a particle in a cell is governed by a transfer function describing the probability of the particle spending a given length of time in the cell. This is based on a 1-D displacement concept within the cell as opposed to a mixing cell concept. Thus, we call this particle-tracking approach the RTTF method. The schematic plot shown in Figure 7-2 illustrates the basis of the RTTF approach. For a cumulative probability distribution function of particle residence times, we compute the residence time of a particle in a cell by generating a random number between 0 and 1 to determine the corresponding residence time from the distribution function. In this example, the advection-dispersion equation was used to generate the RTTF curve, but other transport mechanisms can be incorporated as well, as we demonstrate below. If a large number of particles pass through the cell, the cumulative residence time distribution (RTD) of particles in the cell will be reproduced. Particle-tracking models of single-fracture transport (Yamashita and Kimura 1990) and fracture networks (Robinson 1988) have employed this approach to simulate fracture transport of diffusion into the rock matrix.

The mass of fluid in the computational cell and the mass flow rate to or from each adjacent cell is obtained from the solution of the flow field in a numerical model. In the simplest case, the residence time of a particle in a cell, τ_{part}, is given by

$$\tau_{part} = \tau_f = \frac{M_f}{\sum \dot{m}_{out}} \tag{7-1}$$

where M_f is the fluid mass in the cell and the summation term in the denominator refers to the outlet mass flow rates from the cell to adjacent cells. In the absence of dispersion or other transport mechanisms, the transfer function is a Heaviside function that is unity at the fluid residence time τ_f, because for this simple case, all particles entering the cell will possess this residence time.

7.4.2.1 Sorption

Equilibrium, linear sorption is included by correcting the residence time by a retardation factor. Thus, $\tau_{part} = R_f \tau_f$, and R_f is given by

$$R_f = 1 + \frac{\rho_b K_d}{\phi_f S_f} \tag{7-2}$$

where K_d is the equilibrium sorption coefficient, ρ_b is the bulk rock density of the fracture lining minerals, ϕ_f is the fracture porosity, and S_f is the fracture saturation of the phase in which the particle is traveling. Once again, in the absence of other transport processes, the transfer function is a Heaviside function. As discussed in Section 7.2.4, the linear sorption model is used in such a way as to provide a minimum bound on the influence of sorption (Meijer 1992; pp. 18-19).

Before discussing more complex transfer functions for the RTTF method, we outline the method for determining which cell a particle travels to after completing its stay at a given cell. The assumption that is consistent with the RTTF method is that the probability of traveling to a neighboring cell is proportional to the mass flow rate to that cell. Only outflows are included in this calculation; the probability of traveling to an adjacent node is 0 if fluid flows from that node to the current node. A uniform random number from zero to one is used to make the decision of which node to travel to. Thus, the particle-tracking algorithm is: (1) compute the residence time of a particle at a cell using the RTTF method; and (2) send the particle to an adjacent cell randomly, with the probability of traveling to a given cell proportional to the mass flow rate to that cell.

7.4.2.2 Dispersion

Transport processes such as dispersion can be incorporated into the RTTF particle-tracking algorithm through the use of transfer functions. For dispersion, within a computational cell, we assume that 1-D, axial dispersion is valid. The transport equation and boundary conditions for the 1-D, advective-dispersion equation are

$$R_f \frac{\partial C}{\partial t} = D_{eff} \frac{\partial^2 C}{\partial x^2} - u \frac{\partial C}{\partial x} \tag{7-3}$$

$$C = 0 \quad ; \quad t = 0 \tag{7-4}$$

$$C = C_0 \quad ; \quad x = 0 \tag{7-5}$$

$$C = 0 \quad ; \quad x \to \infty \tag{7-6}$$

where C is the concentration, C_0 is the injection concentration, u is the flow velocity, and D_{eff} is the effective dispersion coefficient, given by $D_{eff} = \alpha u$, where α is the dispersivity of the medium. Here it is assumed that the flow dispersion component of D_{eff} is large compared to the molecular diffusion coefficient D_{AB}. A non-dimensional version of Equation (7-3) can be obtained by the following transformations:

$$\hat{C} = C / C_0 \tag{7-7}$$

$$x = \hat{x} / L \tag{7-8}$$

and

$$\theta = R_f u t / L \tag{7-9}$$

where L is the path length to the point of observation. For the UZ transport problem, L is the distance from the potential repository to the water table. The solution to Equations (7-3) through (7-6) is given by Freeze and Cherry (1979)

$$\hat{C} = \frac{1}{2}\left[erfc\left(\frac{\sqrt{Pe}(1-\theta)}{2\sqrt{\theta}} \right) + e^{Pe} erfc\left(\frac{\sqrt{Pe}(1+\theta)}{2\sqrt{\theta}} \right) \right] \tag{7-10}$$

where Pe is the Peclet number, $Pe = uL/D_{eff} = L/\alpha$.

The use of this solution in the RTTF particle-tracking method requires that the transport problem be advection dominated, such that during the time spent in a computational cell, solute would not tend to spread a significant distance away from that cell. Then, the approximate use of a distribution of times within a single cell should be adequate. Quantitatively, the criterion for applicability is based on the grid Peclet number $Pe_g = \Delta x/\alpha$, where Δx is the smallest characteristic length scale of the computational cell. Note that in contrast to conventional numerical solutions of the advection-dispersion equation, coarse spatial discretization is helpful for satisfying this criterion. Of course, the mesh spacing must be small enough to provide an accurate flow solution. Highly dispersive transport, $Pe_g < 1$, invalidates the assumptions of the RTTF particle-tracking technique. This is not viewed as a significant limitation of the method, because accurate solutions to the advective-dispersion equation are easily obtained by conventional finite-difference or finite-element techniques, when dispersion coefficients are large. For calculations performed for TSPA-VA, α ranged from 7.5 m to 32.5 m, averaging 20 m, while the smallest values of Δx (in the vertical direction) ranged from 18 m to 45 m, averaging 32 m (see Chapter 2, Figure 2-22; grids from the TSw4 to the water table). Furthermore, dispersion has been found to have little influence on UZ radionuclide transport results (see Section 7.6). Therefore, any errors introduced due to the effects of dispersion and grid size are expected to be negligible.

For multi-dimensional flow systems, the dispersion model developed for 1-D systems can be extended to include dispersion coefficient values aligned with the coordinate axes. For this case, the flow direction is determined by the vector drawn from the nodal position of the previous cell to the current cell, and the dispersivity for this flow direction is computed from the equation for an ellipsoid:

$$\alpha = \frac{L}{\sqrt{\Delta x^2 / \alpha_x^{\ 2} + \Delta y^2 / \alpha_y^{\ 2} + \Delta z^2 / \alpha_z^{\ 2}}} \tag{7-11}$$

The RTTF particle-tracking technique cannot be simply formulated with a longitudinal and transverse dispersion coefficient model, with the tensor aligned with the flow direction, because the flow rates between cells are defined rather than the actual flow velocity at a position. For a dispersion model aligned with the flow direction, a random-walk particle-tracking method such as that of Tompson and Gelhar (1990) or a conventional finite-element or finite-difference solution to the CD equation, such as the reactive transport solution module in FEHM, must be used.

The numerical implementation of this technique requires the determination of the dimensionless time θ in Equation (7-9) for a randomly determined value of the dimensionless $\hat{C}$ concentration. This determination is accomplished numerically in the particle-tracking code by fitting Equation (7-10) at selected values of *Pe* between 1 and 1,000 using a piecewise continuous series of straight lines spanning the entire range of $\hat{C}$ values. Then, the value of θ at an arbitrary value of *Pe* is computed by linear interpolation between θ values determined at the Peclet numbers that bracket the actual value. This technique, involving a simple search for the correct type curves, followed by the calculation of two values of θ and an interpolation, is much more computationally efficient (about a factor of two in cpu time) and robust than an iterative approach to the exact solution using Newton's method. Solutions of adequate accuracy (less than 1 percent error) for Peclet numbers between 1 and 1,000 are easily obtained using this linear-interpolation method.

7.4.2.3 Matrix Diffusion

Matrix diffusion has been recognized as an important transport mechanism in fractured porous media (e.g., Neretnicks 1980; Robinson 1994). For many hydrologic flow systems, fluid flow is dominated by fractures because of the orders-of-magnitude larger permeabilities in the fractures compared to the surrounding rock matrix. However, even when fluid in the matrix is completely stagnant, solute can migrate into the matrix via molecular diffusion, resulting in a physical retardation of solute compared to pure fracture transport. Matrix diffusion has been shown to be consistent with measurement on the laboratory scale by Reimus (1995) and on the field scale both by Maloszewski and Zuber (1993) and, at the C-Wells by Reimus and Turin (1997).

To derive a transfer function for matrix diffusion, we must first develop an idealized representation of the transport system. Figure 7-3 shows the geometry of the model system used for this purpose. The geometry and flow system consists of equally spaced, parallel fractures, each of which transmits equal flow. Fluid in the rock matrix is stagnant. Transport in the fractures is governed by Equation (7-3), whereas transport within the matrix is described by the one dimensional diffusion equation

$$R_{f,m} \frac{\partial C}{\partial t} = D_{AB} \frac{\partial^2 C}{\partial y^2} \tag{7-12}$$

where $R_{f,m}$ is the retardation coefficient for the matrix. The molecular diffusion coefficient is the product of the free diffusion coefficient of the solute in water and a tortuosity factor to account for the details of diffusion through a tortuous, fluid-filled pore network. In this model, we treat D_{AB} as the fundamental transport parameter, recognizing that it is a property of both the solute and the medium.

Solutions to this transport problem depend on the nature of the boundary condition in the y-direction. An analytical solution is given by Tang et al. (1981) for the semi-infinite boundary condition:

$$\frac{\partial C}{\partial y} = 0 \; as \; y \rightarrow \infty \tag{7-13}$$

For the case of plug flow (no dispersion) in the fractures, Starr et al. (1985) show that the solution reduces to

$$\frac{C}{C_0} = erfc\left[\frac{\phi \tau_f \sqrt{R_{f,m} D_{AB}}}{b\sqrt{t - R_f \tau_f}}\right] \tag{7-14}$$

for $t > R_f \tau_f$, and $C/C_0 = 0$ for $t <= R_f \tau_f$, where b is the fracture aperture and τ_t is the downstream position at which C is being calculated, divided by the flow velocity in the fractures. The semi-infinite boundary condition between fractures limits the validity of either of these solutions to situations in which the characteristic diffusion distance for the transport problem is small compared to the fracture spacing, S. Robinson (1994) presents a numerical solution to this problem for the boundary condition,

$$\frac{\partial C}{\partial y} = 0 \quad at \quad y = \frac{S}{2} \tag{7-15}$$

The extent of matrix diffusion can be parameterized using the diffusion number,

$$Di = \left(\phi\sqrt{R_{f,m} D_{AB}}\right)/b. \tag{7-16}$$

As long as the solute has insufficient time to diffuse to the centerline between fractures, the solution provided by Tang et al. (1981) can be used as the transfer function for the particle-tracking technique. A sensitivity analysis indicates that values of $Di > 0.1$ invalidate the assumptions of the Tang et al. (1981) solution, while for $Di < 0.1$, the solution is adequate. At higher values of Di, diffusion into the matrix is so pervasive that the semi-infinite boundary condition is not valid. In the extreme case of very high Di, the system reverts to 1-D behavior, with the transport time given by the fracture transport time times the ratio of the total porosity

The RTTF particle-tracking technique developed here does not handle this limiting behavior. Under these conditions, the transport problem should be recast using a continuum model with the matrix porosity used to compute transport times.

An order-of-magnitude estimate shows that this restriction is not limiting for the fracture/matrix flow system at Yucca Mountain. Present-day deep percolation is on the order of 10 mm/yr. The fractures may be assumed to have a porosity of roughly 10^{-4}. The fracture hydraulic conductivity is at least 10^{-7} m/s, (or 3 m/yr) and the fracture van Genuchten β is approximately 2. Assuming gravity-driven flow that is carried primarily by the fracture continuum (at least in the TSw and the CHn zeolitic), the fracture saturation is approximately 0.4. Therefore, the travel time for a distance of 300 m is about 1 year. Note that this travel time is only for a nonreactive solute that travels exclusively through the fractures and is not necessarily representative of actual travel times for real solutes that will interact with the matrix. The diffusion length scale, δ, is approximately given by

$$\delta = \sqrt{D_{AB}\, t}$$

where D_{AB} is the matrix diffusion coefficient, which is about 10^{-10} m^2/s. For a duration, t, of 1 year, the diffusion length δ is about 0.06 m. Given a fracture spacing on the order of meters (see Section 2.1), the assumption concerning diffusion to the centerline between fractures is expected to be valid.

Although in principal the solution of Tang et al. (1981) could be used directly for the transfer function, its complex form makes it very inconvenient for rapidly computing particle residence times. Instead, a two-step process is used wherein the residence time in the fracture is first computed using the transfer function for 1-D dispersion (Equation 7-10) without sorption. This fracture residence time is then used in the plug-flow equation with matrix diffusion and sorption (Equation 7-14) to compute the particle residence time. To use Equation (7-14) as a transfer function, a numerical algorithm was developed to determine the inverse of the complementary error function, that is, the value of x_d for a given value of y_d , such that $y_d = erfc\ (x_d)$. The numerical implementation of this method entails dividing the error function into piecewise continuous segments from which the value of x_d is determined by interpolation. The use of the two-step approach is justified because of the principle of superposition, which allows the dispersive process in the fracture to be decoupled from diffusive transport in the matrix.

7.4.2.4 Radioactive Decay

The particle tracking method may be modified to include radionuclide decay. Simulation of radioactive decay with time-dependent releases of tracer particles becomes computationally burdensome for large numbers of particles. For example, the decay calculation for species i with a decay rate λ :

$$N_j = \sum_{m=1}^{N_i} \{1 - \exp[-\lambda(t - t_m)]\} \tag{7-17}$$

where N_j is the number of particles that have decayed from species i, N_i is the number of particles of species i that have been injected, and t_m is the time at which the m th particle is injected into the system. If 500,000 particles of species i are injected into the system, then at each time step, the number of mathematical operations for decay calculations alone are around 2.5 million. To simulate a time period of 1 million years, the typical calculation requires about 200 time steps. Therefore, the total number of operations will reach 0.5 billion. This suggests that for large-scale simulations, the use of Equation (7-17) is inefficient.

To reduce the computational burden in simulations, the decay calculation in Equation (7-17) is approximated with an integral. Multiplying both sides of Equation (7-17) by dt_m and approximating the summation by an integral from t_1 to t_{Ni}, the average injection time interval between particles, results in:

$$N_j = \left\{ (\tau_2 - \tau_1) + \frac{1}{\lambda}\left[\exp(-\lambda\tau_2) - \exp(-\lambda\tau_1)\right] \right\} / dt \tag{7-18}$$

where $\tau_1 = t - t_{Ni}$ and $\tau_2 = t - t_1$. Using Equation (7-18), reduced the amount of operations within one time step from millions of operations to just 10, which greatly increases the speed of simulations.

The accuracy of the integral approach depends on the number of particles and their release history. In general, the use of more particles increases the accuracy. When the same number of particles is used in simulations, the one with the constant release rate has less error. If the release rate changes with time, we divide the release period into several segments so that within each segment the release rate can be treated as constant. Then, the decay calculations are carried out corresponding to each segment. The accuracy of the approximation (Equation 7-18) is also dependent on the magnitude of the injection time between particles (dt) times the decay constant, λ. If this product is small compared with one, then the approximation is accurate, that is if

$$e^{-\lambda\, dt} \approx 1 - \lambda\, dt. \tag{7-19}$$

Another issue that relates to decay simulations is the spatial distribution of the decayed radionuclides. This problem is treated by using the rule of first in and first out. This rule states that the decay will proceed from the oldest to the youngest particle such that the total number of particles that have decayed equals the number computed in Equation (7-18). This is possible to implement because an injection time is assigned to each particle. This "rule" is not strictly correct, however, because radionuclide decay is a statistical process based on age and cannot be deterministically predicted. To improve this approximation, the radionuclides are assigned to groups based on injection times. The time intervals for each group is based on the time-step selections used for the TSPA-VA calculations. Then the radionuclide decay calculation (Equation 7-18), along with the first in, first out rule, is applied to each injection-time-interval group. In the case where several particles are injected at the same time, a uniform random number generator is used to randomly select the decayed particle(s).

7.4.2.5 Particle Sources and Sinks

There are two methods for introducing particles into the flow system: the particles are either injected with the source fluid entering the model domain or released at a particular cell or set of cells. The first method is used to track source fluid as it passes through the system. The number of particles entering with the source fluid at each cell is proportional to the source flow rate at that cell, which is equivalent to injecting fluid with a constant solute concentration. For method two, an arbitrary number of particles are released at each specified cell, regardless of the source flow rate. In the present study, method one is used for ^{36}Cl and major-ion chemical simulations in which the solute is entering with the infiltrating fluid, and method two is used for radionuclide migration predictions. When fluid exits the model domain at a sink, the model treats this flow as another outlet flow from the cell. The decision of whether the particle leaves the system or travels to an adjacent cell is then made on a probabilistic basis, just as though the fluid sink were another connected cell. Thus, the complexities discussed by Zheng (1994) for handling a so-called weak sink are avoided in the RTTF particle-tracking model.

The base-case model for TSPA-VA uses an automated particle generation scheme. The user selects the total number of particles for each radionuclide, and then FEHM computes the release rate for the particles representing that radionuclide. Particle release for any radionuclide only begins after the source term initiates release of a given radionuclide. At each time step, for each radionuclide, the code checks the number of empty particle slots left in the computer memory and divides by the remaining simulation time (in years). This ratio is multiplied by the duration of the time step to give the number of particles injected at each time step. When the number of injected particles is smaller than the number of injection nodes, FEHM randomly picks injection nodes from the designated injection nodes and randomly selects an injection time within the injection time interval.

A mass conversion factor, corresponding to mass released and the number of particles used at each time step for each radionuclide, is then calculated and recorded for use later on when converting the particles back into mass. Due to variations in input mass and the number of particles used, the conversion factors vary from time step to time step. When passing the mass data back to RIP, FEHM uses the conversion factors and the molecular weight data to convert the particles back into mass. For the simpler cases, including the base-case calculations and most of the sensitivity studies, the particle release rates for a given radionuclide are constant with time (after the source term begins to release that radionuclide). For cases calculated using the chain-decay model including decay-ingrowth, particle release rates are not constant for the parent-daughter species.

7.4.2.6 Transient Flow Fields

Although TSPA-VA calculations have been conducted using the quasi-steady flow approximation described in Section 7.4.4, some sensitivity analyses presented in Section 7.6 do consider transient flow. When the RTTF particle-tracking method is implemented for a time-varying fluid flow system, the approach is somewhat more complex but still tractable. Consider a numerical simulation in which a discrete time step is taken at time and a new fluid flow field is computed. In this model, the new fluid flow time t_{new} is treated as an intermediate time at which the particle-tracking calculation must stop. The fate of all particles is tracked from time t to time

t_{new}, assuming that the flow field is constant over this time interval. When the simulation reaches t_{new}, the position of the particle is recorded, along with its fractional time remaining at the cell and the randomly generated y-coordinate of the transfer function used for that particle in the cell. When the new fluid flow solution is established, the remaining residence time for a particle is determined from the following steps: (1) compute a new fluid residence time t_f; (2) using the y-coordinate of the transfer function previously computed and the new transfer function, calculate a new particle residence time; then (3) multiply this time by the fractional time remaining in the cell to obtain the remaining time in the cell. This method approximates the behavior in a transient system, while reducing to the behavior that would be obtained in an unchanging flow field had the calculation not been forced to stop at the intermediate time.

Another transient effect that must be considered is that the sum of the outlet mass flow rates, $\sum \dot{m}_{out}$ in Equation (7-1) does not necessarily equal the sum of the inlet mass flow rates. When there is net fluid storage in a cell, the particle-tracking algorithm uses the sum of the inlet flow rates in Equation (7-1), whereas Equation (7-1) itself is used when there is net drainage of fluid.

7.4.2.7 Dual-Permeability Formulation

The matrix diffusion transport mechanism developed earlier accounts for the movement of solute into the matrix by molecular diffusion in the absence of advective fluid flow. However, in dual-permeability flow and transport model formulations, fluid flow occurs within each continuum, and a transfer term accounts for fluid flow between the fracture and matrix. Extension of the RTTF particle-tracking technique to handle dual-permeability systems is very straightforward, because the fracture-matrix flow term is simply an additional inlet or outlet flow rate from the node. In the algorithm, this flow term is treated like flow to any other node, and the particle can shift from one continuum to the other. In addition, to simulate molecular diffusion as an additional fracture-matrix transport interaction term, the matrix diffusion option can be invoked for particles traveling in the fracture continuum. Therefore in this formulation, particles that diffuse into the matrix from the fracture do not participate in advective transport in the matrix, while particles that advect into the matrix advect do not diffuse.

7.4.3 Comparison of Particle Tracking and Differential Material Balance Methods

7.4.3.1 Comparison with One-Dimensional Analytic Solutions; Single Porosity System

The first set of simulations are 1-D flow problems for which the dispersion and sorption can be tested against analytical solutions. The flow system is a simple 1-D grid in the direction of flow. A steady-state flow field is established for a ten-node, 1-D grid, after which the RTTF particle-tracking technique is used to simulate the transport.

In the first suite of 1-D tests, a short pulse of particles is injected into the system to test the dispersion transfer function. The responses of these pulse injections of solutes are compared to the time-derivative of Equation (7-10), given by

$$C = \frac{1}{2\theta}\exp\left[\frac{-Pe(1-\theta)^2}{4\theta}\right]\sqrt{\frac{Pe}{\pi\theta}}. \tag{7-20}$$

The equation, which is differentiated, is a step-function; its time-derivative given in Equation 7.4-20 is the solution for a Dirac spike input, which is equivalent to the probability density function for a single atom of the nuclide.

The cumulative residence time distribution (RTD) is also compiled from the particle statistics at the outlet node and compared to Equation (7-10). Figure 7-4 shows that the cumulative RTD solution is reproduced almost exactly for $Pe > 10$, corresponding to a grid Peclet number of 1, as long as a sufficient number of particles is used in the simulation. For this transport system, the $Pe = 20$ results become affected by statistical fluctuations when less than 10,000 particles are used, whereas for 100,000 particles, the breakthrough curve is represented quite accurately, as shown in Figure 7-5. This effect is problem dependent; a sufficient number of particles must be determined empirically for each application of the particle-tracking technique by comparing the results with a simulation using more particles.

Figure 7-4 also shows that for low enough values of Pe, such as the $Pe = 10$ case in this example, the RTTF particle-tracking simulation begins to deviate noticeably from the analytical solution. This case corresponds to a grid Peclet number of 1, which is a practical cut-off below which one cannot obtain accurate *fluid flow* results using this technique. The solution can be made more accurate using a coarser finite element grid, keeping in mind that the restrictions for obtaining an accurate fluid flow solution still exist. The practical implication of this result is that the RTTF particle-tracking technique is most useful for advection-dominated problems (large values of Pe). This will generally be the case for large-scale two- and 3-D problems, which necessarily result in large spatial discretization due to computational limitations.

Finally, the agreement of the sorption curve in Figure 7-4 with the analytical solution shows that linear, equilibrium sorption is also properly accounted for by the RTTF particle-tracking model.

7.4.3.2 Comparison with One-Dimensional Analytic Solutions; Dual Porosity System Including Matrix Diffusion

Figure 7-6 shows a series of simulations of the RTTF particle-tracking algorithm, along with the solution of Tang et al. (1981) for matrix diffusion with and without dispersion in the fractures. The agreement with the analytical solution is very close for either small or moderate amounts of diffusion (curves a and b), for the case of diffusion and sorption on the fracture surface and in the matrix (curve c), and for dispersion, diffusion, and sorption in both media (curve d). This agreement shows that the two-step RTTF approach outlined in the Mathematical Development section provides an excellent approximation of the combined diffusive-dispersive transport system.

7.4.3.3 Comparison with Two-Dimensional, Finite Element Solution for Interwell Tracer Transport

In the previous section, the particle-tracking method was tested in the context of simplified flow fields and geometries, so that analytical solutions or other simple comparisons could be used. In this section, we benchmark the RTTF particle-tracking module against a numerical finite-element solution of the advection-dispersion equation using FEHM for a complex, 2-D flow field. Although this test problem is for a saturated system, it serves as an appropriate test of the numerical method of incorporating hydrodynamic dispersion into the particle tracking method.

Figure 7-7 is a plan view of the model domain and grid. This simulation is designed after the field tracer experiments conducted at the C-Wells (Robinson, 1994). These tests are designed to measure the hydrologic and transport properties of the SZ beneath Yucca Mountain. A lognormal permeability field was generated with a Gaussian spatial correlation structure. The mean of the distribution is 10^{-12} m^2, standard deviation is 1 (units of $\ln(m^2)$), and a correlation length is 20 m. The permeabilities and porosity (10^{-4} at all positions) are chosen to represent a hydrologic system dominated by fracture flow. A steady-state flow field is established between the injection and production wells (the filled circles in the figure) at a flow rate of 1.58 kg/s, after which time a slug of tracer is injected, and the inlet fluid and concentration versus time at the production well is recorded.

Figure 7-8 shows the results of the finite-element calculation and two particle-tracking simulations discussed below. The breakthrough curve is characteristic of field tracer experiments in fractured porous media. The heterogeneous permeability field and the point source and sink inlet and outlet give rise to a broad distribution of breakthrough times, long tailing of the breakthrough curve, and less than 100 percent recovery of tracer at the time when the experiment would typically be terminated. The RTTF particle-tracking simulations assume dispersivity values of 0 and 1 m. The finite element solution (dispersivity of 1 m) exhibits some numerical dispersion that is not present in the particle tracking solution. Nonetheless, the finite-element and particle-tracking curves exhibit virtually identical peak times and mass recovery versus time curves. The particle-tracking solution with no dispersion exhibits noise that will be present no matter how many particles are used. The noise is due to the fact that there are a finite number of pathways between the injection and production points. With the RTTF particle-tracking technique in the absence of dispersion, a particle in a particular path will always possess exactly the same transit time as others in that path. Therefore, the breakthrough curve reflects the arrival of these discrete paths as "bursts" of tracer, resulting in noise in the computed curve and a higher peak concentration. However, this is not viewed as a limitation, since a dispersivity value typical of those estimated in field transport studies is large enough to eliminate the noise.

The comparison of the particle-tracking and finite-element solutions suggests that the particle-tracking algorithm can duplicate the behavior of a system exhibiting macrodispersivity in which the dispersion is due to heterogeneity in the permeability field. Furthermore, the particle-tracking solution eliminates the numerical dispersion present in the finite-element solution, which results in an underestimation in the first arrival time, a parameter often reported in interwell tracer studies. Finally, the exceptional computational efficiency of the technique makes it a logical choice in stochastic analyses requiring multiple realizations. For this example, this

100,000-particle simulation required only 5 percent of the cpu time of the advection-dispersion equation solution.

7.4.4 Model Abstraction for Long-Term Transient Flow

Most flow and transport calculations of the UZ have assumed steady flow conditions. There is good reason to believe that short-term transients at the time scale of individual precipitation events are damped in the nonwelded PTn unit, so that the flow field in the underlying units experience a much smoother input of flux than what occurs at the surface. This is examined in greater detail in Section 7.6.4 where sensitivity of this assumption to parameter sets and PTn thickness is evaluated. Fundamentally, the reason short-term transients are damped in the PTn is because the matrix permeability of the PTn is of the same magnitude as the fracture permeability (see Chapter 2). Therefore, there is no reason for high flow rates to preferentially select the fracture continuum over the matrix continuum. In fact, the matrix continuum will remove water from the fracture continuum due to the stronger capillary pressures that evolve in the matrix as compared with the fractures given the same level of effective permeability. With regard to longer term transients such as climate-change induced variations in percolation flux, there has been considerably less effort devoted to determining the influence of such events and the validity of the steady state assumption for computing flow distributions and for making transport predictions. The practical reasons for considering steady flow as the starting point is that the computational burden of transient fluid flow and solute transport calculations is much greater. Furthermore, it could be argued that if a simpler steady quasi-steady flow and transport model does a good job at explaining the data (i.e., pore water and perched water chemistry) at Yucca Mountain, it should be favored over a more complex, transient model.

Even if steady models prove to be sufficient for explaining present-day conditions, there is a need to compute the impact of long-term transients on the ability of the UZ to impede the transport of radionuclides. There is ample evidence in the record of long-term variations in climate, which in turn led to changes in percolation rate and flow characteristics. Earlier we showed that percolation rate is a controlling parameter on radionuclide transport, so capturing the changes likely to occur in the future is important. The method used in TSPA-VA analyses to capture long-term transients is a quasi-steady flow and transport model in which the flow is assumed to jump instantaneously from one steady flow field to another. For this type of model to be valid, we must first show, through transient analyses, that the simplifying assumption does not significantly change the results. Furthermore, to provide added confidence, we must demonstrate an understanding of the time scales and magnitudes of the changes occurring in the transient simulation. In the next section we present an analysis of flow and transport in a transient system is presented that provide the basis for making the simplifying assumptions just discussed.

The following analyses (and others as identified in this chapter) use the hydrogeologic parameter sets developed by Bodvarrson et al. 1997 for UZ flow calculations. The parameter sets given in this reference were developed based on the same inverse modeling techniques described in Chapter 2, Section 2.4.3.1, which was used to develop the base-case hydrogeologic parameter sets (Tables 2-18 through 2-22). The parameter sets are close to, but not exactly the same as, the base-case hydrogeologic parameter sets identified in Chapter 2. The Bodvarrson et al. (1997)

parameter sets were used for some of the UZ transport calculations in this chapter because they were available at an earlier date than the base-case parameter set.

7.4.4.1 Comparison of Process Model and Abstraction

Hydrologic Response to Transient Infiltration - To develop a model for long-term transient flow and transport, we must first understand the magnitude and time scale of changes in the flow field caused by long-term changes in the percolation rate. The three cases considered are: Case 1—increase in percolation rate from the base map to the 3 × I case; Case 2—decrease in percolation rate from the base map to the I/3 case; and Case 3—increase in percolation rate from the I/3 case to the base map, where the "I" case is given by Flint et al. 1996, and is the present-day, expected value infiltration condition. The I*3 and 1/3 are the spatially varying infiltration rates multiplied and divided by three, respectively. Hydrologic parameter set 6412b (Bodvarrson et al. 1997) was chosen to perform this analysis. Figure 7-9 shows the water flow rate (computed using the FEHM module for unsaturated flow) exiting the 2-D model domain versus time for the three cases. The fluid flow response at the water table in response to a flux increase begins to be seen after only about 50 to 200 years, depending on the absolute level of the starting infiltration rate. This rapid response is due to flow through permeable fracture pathways, which, because of their low storativity, take very little time to adjust to the flow rate increase. The ramp to the eventual water mass flow rate takes longer, from 500 to 5,000 years. There is a very gradual rise to the plateau values at long times, due to gradual increase in matrix saturation values. The time scale for that process is governed by the fracture/matrix interaction parameter. A discussion of what the fracture/matrix interaction parameter represents and how it is established is given in Chapter 2, Section 2.4.4.3. The infiltration rate decrease scenario (Case 3) exhibits similar time scales as Case 2, the increase between these same two infiltration values.

The matrix saturations for four locations below the crest of Yucca Mountain are shown in Figures 7-10 (tsw5 and bf2z) and 7-11 (ptn3 and ch4v). Two effects are giving rise to the transient response. First, the infiltration changes are felt first in the units closest to the ground surface, as expected. However, in addition to the transient reaching a given location, the rate of imbibition or drainage between the fractures and matrix is a strong function of the fracture/matrix interaction term. In a welded unit such as tsw5, fluid transfer is slow because of the large reduction factor for the fracture/matrix interaction term. Therefore, rock in this unit responds to changes in percolation rate more slowly than the underlying ch4v and bf2z units. The time scale for the changes to take place in the matrix saturations of tsw5 are between 1 and 10 ky, depending on the infiltration scenario. This result provides evidence that if the climate last changed about 10 ky ago, the matrix saturations have probably had enough time to equilibrate to their new saturation values, rather than being reflective of a past climate scenario. For the welded units, this conclusion can be made with less confidence than for the nonwelded units, given the different time scales involved. Finally, note that the absolute magnitude of the changes in matrix saturation are quite small. This result shows that it is acceptable to use hydrologic parameter values from parameter estimation simulations at one infiltration rate for simulations at other fluxes within a factor of three of the original value without significantly affecting the fit to the matrix saturation data.

Neptunium Transport in a Transient Flow Environment - The nature of radionuclide transport during a long-term transient is examined using FEHM for flow and transport with the

following scenario. The radionuclide ^{237}Np is simulated with K_d = 2 ml/g in the zeolitic tuff, with a constant release rate from the near-field environment from 1 ky to 31 ky, after which it was assumed that the entire inventory was depleted. At 5 ky, we assume a climate-change-driven increase in the infiltration rate from the I/3 map to the base (I) map, after which the flux remains constant for the duration of the simulation. Particle tracking simulations in fully transient mode were used to simulate the radionuclide breakthrough curve at the water table. This result is shown in Figure 7-12, along with other simulation results used for comparison. The radionuclide mass flux in the transient simulation tracks the base-map result exactly for the first 5 ky, rises sharply as the repository horizon flux responds to the higher infiltration, and then tracks the I result closely after about 10 ky. The rise in the flux value to levels greater than the upper steady state curve is the result of the radionuclide mass resident in the flowing water at the time the flow rate increase occurs. Since the release rate is assumed constant despite the change in flow rate for this simulation, the concentration within the fluid at the time of the flow rate change is greater than values thereafter. The higher infiltration rate carries this solution to the water table at a greater rate, thereby temporarily increasing the mass flux to levels greater than even the I steady state curve. Once the concentrated fluid is flushed from the UZ, the breakthrough curve reverts to the curve predicted for the higher infiltration rate case. Note that if a more realistic scenario of infiltration-dependent release rate were simulated, the result presented here would probably be masked the increased radionuclide flux from the repository. Nevertheless, the simulation shows that except for the transient effect, the flux at the water table is closely approximated by the appropriate steady state curve (I/3 curve before 5 ky, I thereafter).

The final comparison in this transient analysis is to examine the quasi-steady assumption, comparing it to the full transient simulation. As shown previously in Figure 7-12, the agreement is extremely good, even during the transient period. The differences are shown on an expanded scale in Figure 7-13. The quasi-steady assumption was run assuming that the new, high-flux flow field took effect exactly at 5 ky. Instead of the finite time required to reach the steady state at the higher percolation flux, the influence of the transient appears immediately at the water table. However, as shown, the flow field achieves a steady state at a higher infiltration rate within 100 to 1,000 years, a very short time interval in a simulation of this scale. This is the reason that the quasi-steady assumption works as well as it does. Although further testing with different parameter sets, radionuclides, and near-field environment release rates should be carried out, it appears that the quasi-steady assumption is an excellent approximation to the full transient solution.

7.4.4.2 Effects of Water Table Rise on Radionuclide Transport

One other climate-related effect that must be considered is the impact of a rise in the water table due to a future wetter climate. For the UZ model, the elevation of the water table is set as an imposed boundary condition, and thus is not a computed result. Estimates based on mineralogical studies and SZ modeling efforts have placed the water table as roughly 100 m above the present-day elevation. In this model, we can easily assign the water table a higher elevation by specifying all nodes below a certain elevation as water table nodes. In a sensitivity study of water table elevation, we assume a water table elevation of 830 m, and perform the simulation with the base infiltration map and parameter set 6541 (Bodvarrson et al. 1997). Figures 7-14 and 7-15 are the resulting Green's function (response to a pulse input) and cumulative RTD curves for this system. As can be seen in the figures, arrival times at the water

table are somewhat higher for the elevated water table compared to the present-day water table position, as expected. However the differences are not large. The higher water table places more of the CHn and underlying units under the water table. This includes some of the zeolitic units that provide retardation of radionuclides.

The effects of water table rise in the TSPA-VA calculations will be included for the climate states known as "LTA" and "super pluvial" (see Section 2.4.1.1). For the LTA climate, for which the mean infiltration is enhanced over the present day climate, the water table is assumed to instantaneously rise 80 m at the onset of the climate change. For the super pluvial climate, which has the highest mean infiltration of the three climate conditions, the water table is assumed to instantaneously rise 120 m at the onset of the climate change. The instantaneous change in water table position means that when the climate changes to a higher infiltration and water table position (e.g., present-day climate to LTA climate), radionuclides that were in the UZ at elevations between the water table positions for the two climates will be immediately available for transport in the SZ. This leads to the generation of small pulses of radionuclide mass flux in the SZ upon such climate changes. For the opposite type of climate change, the water table position drops, and there will be a brief period for which there will be a drop in the radionuclide mass flux at the water table as the radionuclides advance into the portion of the UZ that became available with the climate change.

7.4.5 Colloid-Facilitated Radionuclide Transport

One conclusion from the UZ Radionuclide Transport Abstraction/Testing workshop (see Section 7.3) is that a credible model for plutonium movement in the UZ must include transport of Pu radiocolloids and "pseudo-colloids" (Pu attached to natural colloidal material or colloids introduced in the water due to construction of the repository) must be considered. To date, process-level transport models and performance assessment models have assumed that plutonium migrates as an aqueous component. Since sorption coefficients of aqueous plutonium on Yucca Mountain tuffs are large, plutonium has been predicted to migrate only a short distance before undergoing radioactive decay. Colloid-facilitated transport has the potential to increase transport velocities considerably, and thus is an important addition to the TSPA calculation.

In Section 7.4.5.1 derives an analytical solution for 1-D transport in a dual-porosity system for the case of radionuclides that are reversibly sorbed on colloids, present results of the model, and make a first attempt to quantify how different model assumptions and chemical parameters will impact the predicted transport of radionuclides that reversibly sorb on mobile colloids. This model is then incorporated into the TSPA-VA transport model to predict plutonium migration. Other radionuclides may also migrate in colloidal form, (such as Americium), however for the base-case set of nine radionuclides, Pu is expected to be the most likely candidate for colloidally-facilitated transport (see Chapter 6, Section 6.4.2.1).

In Section 7.4.5.2 presents a model for treating transport of radionuclides that are irreversibly sorbed to colloids. In this case, the radionuclides are permanently attached to colloids at the point of generation within the engineered barrier system and remain attached to colloids for the duration of the transport through the geosphere.

7.4.5.1 Model Abstraction for Colloid-Facilitated Radionuclide Transport for Reversibly Sorbed Radionuclides

Dual Porosity Model for Transport of Dissolved Aqueous Species in Fractured Rock - For simplicity, the model developed herein is a 1-D transport model of a contaminant travelling in a fractured, dual porosity medium. A dual porosity model allows flow in the fractures only (water in the matrix is stagnant), but dissolved substances do move between the fractures and the matrix by diffusive transport. Before using this model for the TSPA-VA particle tracking routine, the results must be extended to account for 3-D transport and for flow exchange between fractures and matrix. This report will show that the model may be easily extended to 3 dimensions for implementation in the particle tracking transport model coupled with RIP The model is also extended to included colloid migration in the matrix continuum to account for advective exchange of water between fractures and matrix that occurs in the dual permeability formulation. Although colloids may be hindered from entering the matrix of the lower-permeability units due to size exclusion, it may be possible for colloids to migrate through the more permeable matrix characteristic of the CHnv. It is in this hydrogeologic unit where matrix flow is expected to exceed fracture flow (see Chapter 2)

It is assumed that the hydrologic conditions are such that uninterrupted fracture flow pathways exist from the repository to the water table. Thus the model is an "end-member" that attempts to capture the worst-case conditions from the standpoint of colloid-facilitated transport. It must be stressed that the model would severely underestimate the retardation capacity of the UZ to radionuclides, such as plutonium, if it can be demonstrated that rock strata between the repository and the water table exhibit matrix-dominated flow and transport. This concern is demonstrated in the site-scale model calculations that follow. Nevertheless, it is a starting point for addressing performance under conditions of fracture-dominated transport, which is considered a likely scenario.

Aqueous transport: a model available in the literature for the 1-D transport of an aqueous contaminant, and later revise it to include colloidal transport. This first model was developed by Tang et al. (1981). The model geometry is one of transport in a parallel-plate fracture, with the surrounding matrix block on either side of the fracture (Figure 7-16). The transport equation to be solved in the fracture assumes the 1-D advective-dispersion equation, diffusion into the porous rock matrix, equilibrium, and linear sorption on the fracture surface:

$$R_f \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - \frac{q}{b} \tag{7-21}$$

where C is the concentration, t is time, z is the coordinate in the direction of fluid flow, R_f is the sorption retardation factor for the contaminant on the fracture, D_z is the axial dispersion coefficient, v is the fluid velocity, q is the diffusive loss term for the porous rock matrix, and b is the fracture aperture. Assuming no advective flow of fluid in the matrix, the transport equation for the contaminant is

$$R_m \frac{\partial c}{\partial t} = D_m \frac{\partial^2 c}{\partial y^2} \tag{7.4-22}$$

where c (lower case) is the concentration of contaminant in the matrix, y is the direction perpendicular to the flow direction, R_m is the retardation factor for the matrix, and D_m is the effective diffusion coefficient in the matrix rock. These two transport equations are coupled through the diffusive loss term q, formulated by assuming continuity of concentration for fluid in the fracture with that in the matrix at $y = b$, such that

$$q = -\theta D_m \left.\frac{\partial c}{\partial y}\right|_{y=b} \tag{7-23}$$

where θ is the volumetric water content in the matrix (equal to porosity for a saturated matrix, or porosity times saturation for an unsaturated matrix). Tang et al. (1981) derived a full transient solution to this system of equations for any position in the fracture or matrix. Here, we give the result for a position $z = L$ in the fracture, presumed to be at the water table:

$$\frac{C}{C_{in}} = \frac{2\exp(Pe/2)}{\pi^{1/2}} \int_l^\infty \exp\left[-\xi^2 - \frac{Pe^2}{16\xi^2}\right] \operatorname{erfc}\left(\frac{Pe\,A}{4\xi^2\sqrt{\frac{t}{\tau} - \frac{Pe\,R_f}{2\xi^2}}}\right) d\xi \tag{7-24}$$

where C_{in} is the concentration at the entrance to the model, ξ is the integration variable, τ is the mean residence time of fluid through the fracture, equal to L/v,

$$l = \left(\frac{\tau\, Pe\, R_f}{4t}\right)^{1/2} \tag{7-25}$$

and

$$A = \frac{\theta\left(\tau R_m D_{mol}\right)^{1/2}}{2b}. \tag{7-26}$$

The relation between D_z and the dispersivity, α, is $D_z = \alpha v$, from which it follows that the axial dispersion Peclet number $Pe = L/\alpha$. Additional simplifications are possible if axial dispersion is considered to be negligible. Starr et al. (1985) derived a similar model for this case, showing that

$$\frac{C}{C_{in}} = \operatorname{erfc}\left(\frac{A}{\sqrt{t/\tau - R_f}}\right) \quad for \quad t > R_f\,\tau \tag{7-27}$$

with $C/C_{in} = 0$ for $t <= R_f\,\tau$. Although the finite dispersion model, Equation (7-24), will be used for the simulations presented below, Equation (7-27) provides a useful simplification to examine the dependencies in the model. The main parameters are the fracture retardation factor R_f, which

provides a sorptive delay in the breakthrough times from that of water flow in the fracture, and the dimensionless group A, which can be used to assess the relative importance of advective transport in the fracture to diffusion in the rock matrix. High values of A lead to the case in which interaction of contaminant with the rock matrix is important, and would result from any of the following conditions: high matrix porosity, large water travel times in the fracture, high matrix diffusion coefficient, large matrix sorption coefficient, or small aperture.

Coupling with Colloid-Facilitated Transport using Linear Sorption - This section includes a simplified colloidal transport model equation to derive a combined aqueous/colloidal transport model. The key assumptions of the colloidal transport sub-model are that the contaminant sorbs reversibly to the colloids in addition to interacting with the rock matrix and fractures, the colloids are isolated to the fracture, and possess dispersive properties equivalent to that of an aqueous solute, and the colloids interact reversibly with the fracture surface.

Given these assumptions, a transport equation for contaminant attached to colloids can be written as

$$R_c \frac{\partial C_c}{\partial t} = D_z \frac{\partial^2 C_c}{\partial z^2} - v \frac{\partial C_c}{\partial z} \tag{7-28}$$

where C_c is the concentration of contaminant attached to colloids, and R_c is a retardation factor that captures the details of colloid attachment/detachment and filtration processes.

To couple the transport equation for contaminant on colloids (Equation [7-28]) to the aqueous transport equation (Equation [7-21]), reversible, linear sorption of contaminant onto colloids is assumed:

$$C_c = K_c C \tag{7-29}$$

where K_c is the distribution parameter relating the concentrations. Since both C and C_c are expressed as moles of contaminant per unit fluid volume, K_c is a dimensionless parameter expressing the ratio of contaminant mass residing on colloids to the mass present in aqueous form. In terms of more commonly defined and measured quantities, K_c is the product of the distribution coefficient for contaminant on colloids and the concentration of colloidal material available for sorption (mass of colloids per unit fluid volume). Adding Equations (7-21) and (7-28), and making use of Equation (7-29), we obtain for the concentration of the contaminant in the aqueous phase for the fracture:

$$\left(\frac{R_f + K_c R_c}{1 + K_c} \right) \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - \frac{q}{b(1 + K_c)} \tag{7-30}$$

The coupling between the fracture and matrix is identical to the case without colloids, since we assume no mobility of colloids into the matrix rock. Inspection of Equations (7-30) and (7-21) reveals that the forms of the transport equations are identical, with different constants in the accumulation and diffusive loss terms. Therefore, the solutions developed thus far (Equations (7-24) and (7-27) need only be modified slightly to include colloid contaminant transport. To do this, the following revised constants $\hat{R}_f$ and $\hat{A}$ are defined:

$$\hat{R}_f = \frac{R_f + K_c R_c}{1 + K_c} \tag{7-31}$$

$$\hat{A} = \frac{\theta(\tau R_m D_{mol})^{1/2}}{2b(1 + K_c)} \quad . \tag{7-32}$$

Using these expressions instead of their counterparts R_f and A allow us to consider colloidal transport under dispersive conditions (Equation [7-24]) or plug-flow fracture conditions (Equation [7-27]). One final note is that the inlet concentration C_{in} must include contributions from both colloidal and aqueous contaminant, and therefore should be considered to be a total mass of contaminant (aqueous and colloidal) per unit fluid volume.

Implementation in TSPA-V - One can now discuss extension of this model to three-dimensions. The derivation presented, leading up to Equations (7-30), (7-31), and (7-32), could have been done for 3-D transport. Terms in equations involving first and second spatial derivatives in z may be generalized to gradients and Laplacians, respectively, for three dimensions. The flow velocity is a vector in three-dimensions, and in the transport equation is combined as a scalar product with the spatial concentration gradient. The dispersion coefficient in the present model is constant and is independent of direction (the particle tracking method is limited to isotropic dispersion); therefore, an appropriate value is used for D_z for three dimensions as presented in Section 7.4.1.2, subsection on dispersion. None of these changes affects the final results in Equations (7-30), (7-31), and (7–32). Note that the 1-D solution is still needed (Equation [7-24]) to use for residence time calculations as discussed in Section 7.4.1.2, subsection on matrix diffusion.

It is assumed that the colloids do not diffuse, therefore, fracture transport of colloids do not exchange with the matrix. This is an assumption used to simplify the solution and partly relies on the fact that colloid diffusion is small compared with diffusion of molecular species. It is also a conservative assumption in that it tends to promote transport of colloids through the fractures. For a dual-permeability model, water flows through the matrix and between the fractures and matrix, unlike the dual-porosity model used for the development of the colloid transport model. To extend the model to include this effect, the advective exchange water between fractures and matrix is assumed to carry whatever colloids are present. Therefore, colloids can enter and transport through the matrix. This assumption requires a method to treat the transport of colloids through the matrix, in addition to the model developed for transport through the fractures. This

may be easily done with some simple modifications to the fracture transport model. For matrix colloid transport, Equation (7-30) is used with the following modifications: $R_f \rightarrow R_m$, where R_m is the matrix retardation factor for the aqueous species; $q = 0$ (no matrix diffusion); and use values of K_c, R_c, D_z, and v appropriate for matrix transport.

7.4.5.2 Model Abstraction for Colloid-Facilitated Radionuclide Transport for Irreversibly Sorbed Radionuclides

The transport model for radionuclides irreversibly sorbed to colloids is a simple extension to the model for radionuclides that are reversibly sorbed to colloids. Irreversible sorption may be accounted for by multiplying Equation (7-30) by K_c and then letting $K_c \rightarrow \infty$. Then Equation (7-30) becomes

$$R_c \frac{\partial C_c}{\partial t} = D_z \frac{\partial^2 C_c}{\partial z^2} - v \frac{\partial C_c}{\partial z} \quad . \tag{7-33}$$

This is the same equation as previously derived for radionuclide transport through a fracture attached to colloids (Equation [7-27]). The difference here is that the aqueous radionuclide concentration, C, is zero but the concentration of radionuclide irreversibly sorbed on colloids, $C_c = K_c C$ is nonzero as $K_c \rightarrow \infty$.

7.4.5.3 Effects of Colloid-Facilitated Radionuclide Transport

This section examines the potential role of colloids in facilitating transport of strongly sorbing radionuclides such as plutonium. Although the hydrologic and transport model assumptions are simplistic, the analysis places the relative importance of colloids and aqueous radionuclides in perspective by examining the case for which colloid-facilitated transport is deemed to be most problematic, namely fracture flow and transport. For this situation, must still consider aqueous transport processes that serve to retard the movement of radionuclides, such as matrix diffusion and sorption on fractures and in the matrix. Not doing so will lead to poor predictions of repository performance due to the overly conservative assumption that colloids result in a direct, rapid migration to the water table. Although this may be the case for irreversibly-sorbed radionuclides that form stable radiocolloids, some radionuclide/colloid interactions are expected to be less stable, leading to reversible radionuclide/colloid interactions.

The importance of colloid-facilitated transport is discussed with respect to the movement of aqueous radionuclide through the fractured tuff, so that the primary results are presented in terms of possible degradation in repository performance from strictly dissolved, aqueous transport. Colloid-facilitated transport is expected to severely reduce travel times and apparent dispersion when the fraction of radionuclide residing on colloids approaches that present as an aqueous solute. Colloids reduce the diffusive loss of radionuclide to the rock matrix by preventing radionuclide from leaving the fracture via diffusion and sorption to the rock matrix. Furthermore, if sorption onto fractures is important, colloids reduce the effective retardation factor in a manner analogous to aqueous speciation effects in which nonsorbing complexes form at the expense of sorbing complexes. Conversely, fracture sorption can be considered a transport process that "competes with" colloid-facilitated transport if the colloid distribution parameter K_c

is of order 1. Thus the key parameter needed to constrain this problem of colloid transport is K_c, a parameter controlled by the sorption coefficient of radionuclide and the amount of colloidal material present.

Although the model is idealized, it can be used quite simply as a building block of a more sophisticated hydrologic model. The model developed here assumes fracture flow and transport for the fracture domain, with interaction with the surrounding matrix. In the matrix, the sorption coefficient of aqueous plutonium is assumed to be reduced by a factor of $1 + K_c$, and there is no filtration of colloids in the matrix even for advective transport through high-permeability units like the vitric Calico Hills unit. Thus the model is conservative in its assumptions on the migration of colloids through the UZ.

7.5 PROCESS BASE CASE

As discussed in Section 7.1.3.1, the process-level UZ transport code, FEHM is used directly for TSPA calculations of radionuclide transport through the UZ. Therefore, no prior or abstracted calculations of UZ radionuclide transport are needed for use in the TSPA calculations. Due to this approach, there are no process base case calculations to report for UZ radionuclide transport.

7.5.1 Coupling of FEHM Particle Tracking with RIP

For the UZ transport, the 3-D site-scale transport model (Robinson et al. 1996) as implemented by the FEHM particle tracker module (LANL 1997a) is directly called by the total system performance assessment code, RIP (Golder 1996). The UZ flow fields required by the FEHM module are called separately by RIP (see below). Thus, during a given realization, for every simulation time step in RIP (e.g., 100 years) source term information (specifically, radionuclide mass fluxes) is passed from the RIP EBS cells to the FEHM particle tracker, which then moves the particles as far as they would travel during that time step, based on the steady state velocity field in the fractures and matrix of the various UZ rock units. The particle generation scheme for the repository source term is discussed in Section 7.4.2.5. RIP also passes the stochastic UZ transport parameter set for each RIP realization. These stochastic parameter sets are generated using RIP prior to multiple-realization TSPA-VA calculations. Further details on the RIP generation scheme for the transport parameters are given in Section 11.2. The transport parameters that are sampled stochastically are matrix diffusion, matrix sorption, dispersivity, fracture aperture, plutonium aqueous/colloid partition coefficient. The quantitative distributions used for these coefficients are discussed below. FEHM passes back to RIP the mass flux that leaves the UZ model domain in FEHM (i.e., the mass to be passed to the saturated-zone model). This is the mass that enters the water table. The radionuclide mass in the fractures is passed separately from the mass in the matrix, in order to plot intermediate results. These two masses are then mixed in a RIP cell and passed to the SZ convolution integral function.

In this implementation for TSPA-VA, FEHM does not calculate the flow field dynamically, only the transport. The flow fields are pre-generated with TOUGH2 (Pruess 1991; Bodvarsson et al. 1997). This approach, using a finite set of pre-generated flow fields for UZ transport, implies the assumption of quasi-steady flow (see Section 7.4.2). There are a library of flow fields corresponding to the uncertainty in the UZ flow parameters and infiltration rate. For each realization, a different flow field is sampled based on a sampling of the key uncertain parameters

responsible for that flow field. Also for each sampled flow field corresponding to present day climatic conditions, there are two corresponding flow fields that have a higher infiltration flux boundary condition at the surface based on two future climate scenarios, LTA and super-pluvial. A detailed discussion of the UZ flow field, climate states, and generation methods is given in Chapter 2.

7.5.1.1 Interfaces with Repository Source Term and Saturated Zone Radionuclide Transport

Based on the description of repository regions in Chapter 3, the radionuclide source term for the UZ transport model is divided into six distinct regions (based on heterogeneity of infiltration flux, see Figure 7-17a). Thus the there are six distinct sets of EBS cells in RIP which are fed to six distinct areas of the 3-D UZ domain modeled by the UZ particle tracker. Radionuclide release within each of the six regions is defined by the radionuclide mass releases as computed for the EBS. These releases are assigned to the common grid structure that the flow model and transport model employ. The releases within each region are spread uniformly over the grid cells contained within the region. Only a small number of waste packages are expected to release radionuclides over the "early" repository period (the first ten thousand years). For these initial releases from the repository, spreading the relative sparse releases across the potential repository block across the entire repository zone where a given release occurs introduces a large, artificial, dilution of the release. This leads to radionuclide concentrations at the water table that are representative of the average, but lower than the peak concentrations expected. In the later repository period when many waste package are releasing radionuclides, the distribution of releases across the repository zones is believed to be more representative. Despite these effects on radionuclide concentration, the radionuclide mass flux at the water table is accurately represented because the mass flux is not sensitive to radionuclide concentration.

The output of the particle tracker for the UZ has also been divided into six distinct regions at the water table, based in part on the formation lithology intersecting the water table. (The six regions at the water table do not have the same x,y coordinates as the six regions at the repository horizon.) Radionuclide mass flux through each water table region is given as input to the SZ transport model. As mentioned above, fracture and matrix mass fluxes from these six distinct regions are fed back to six RIP mixing cells and then passed to the external function that it is the SZ convolution integral. Both solute and colloid mass are tracked throughout the models.

7.5.2 Example of RIP/FEHM Coupling Radionuclide Transport Calculations

We have calculated radionuclide transport through the UZ using the coupled RIP/FEHM coupled transport model to provide a demonstration of the calculation method for TSPA-VA. Transport is calculated using 3-D particle tracking (see Section 7.4) and the base-case, LTA, UZ flow field described in Section 2.5. In these example calculations, hypothetical tracer particles are released from each of the six repository regions shown in Figure 7-17a. Transport calculations are performed for tracer particles released from each of the six repository regions (NW is north west; NE is north east; SE is south east; SC is south central; SW is south west; CC is central central). The flux of tracer particles at the water table are captured and plotted for the six SZ regions shown in Figure 7-17b. The calculations use a spatially homogeneous source of 500,000

particles within each the six repository regions, issued uniformly over a time period between 5,000 years and 10,000 years after waste emplacement. The source strength in the different repository zones range from 3.45 Ci/yr to 14.95 Ci/yr due the different emplacement areas in the different zones. The total source strength for all six regions is 50 Ci/yr. The transport properties used were the average, base-case values for a nonsorbing radionuclide (i.e., no matrix sorption, matrix diffusion of 3.2×10^{-11} m^2/s, dispersivity of 20 m, fracture aperture of 10^{-4} m, and no colloid partitioning).

In this example calculation, as for the TSPA-VA calculations, the number of particles required are empirically determined through testing of the particular calculation. The (qualitative) criterion used to evaluate the number of particles needed is a comparison of the amount of "noise" in the calculated mass flow rate at the water table to the perceived "signal." Noise in the calculated mass flow rate is apparent in Figures 7-18 through 7-24, and is more pronounced at lower mass flow rates. In this example calculation, the lower mass flow rates correspond to lower numbers of particles, which results in more noise in the calculated mass flow rate. Greater amounts of noise at the lower mass flow rates are tolerable, because the lower mass flow rates are not important for establishing peak radionuclide concentrations at the accessible environment. As it turns out, more particles are required as the simulation time period increases. Therefore, a million-year simulation will require more particles than a one-hundred-thousand-year simulation. Further details on the number of particles used for TSPA-VA calculations are given in Chapter 11.

The responses are shown in Figures 7-18 through 7-23. These figures may be used as a rough guide to the transport pathways through the UZ. In Figure 7-18, the releases from the NW region are primarily found to enter the SZ regions 5 and 6. From Figure 7-17, it is seen that releases from the NW repository region must move laterally to terminate in the water table region 5. Thus, we may interpret these results to be an indication of lateral diversion from west to east for radionuclides released at this location in the repository. The predominant capture of releases from the NE region of the repository in water table region 5, shown in Figure 7-19, also suggests some lateral diversion east and perhaps south. Releases from the CC region of the repository are found in SZ regions 3, 4, and 5 (Figure 7-20). The releases entering water table region 5 from the CC repository region are a clear indication of lateral diversion. Capture of releases from the SE repository region (Figure 7-21) predominately in SZ regions 5 and 2, from the SC repository region (Figure 7-22) predominately in SZ regions 1 and 2, and from the SW repository region (Figure 7-23) predominately in SZ regions 1 and 3 suggests a more vertical, downward pathway at these locations. Thus, the general picture that emerges is that lateral diversion is occurring for releases from the northern portion of the repository, while releases from the southern portion are primarily moving downward. This picture is further supported by a more detailed analysis of transport pathways given in Section 7.6.1.2.7.

The total release to the SZ from the total EBS release is shown in Figure 7–24. The cumulative response at the water table is similar in character to the responses for release in the individual repository zones, with amplification in magnitude due to the larger source.

7.5.3 Inputs For TSPA-VA Unsaturated-Zone Radionuclide Transport

7.5.3.1 Unsaturated-Zone Flow Fields for Radionuclide Transport Calculations

The YMP 3-D, site-scale flow model for the UZ (Bodvarsson et al. 1997; CRWMS M&O 1998, Chapter 2) was used in calculating mountain-scale, unsaturated-zone flow for this TSPA. 3-D modeling was needed primarily because of flow characteristics below the repository, where significant lateral flow is believed to occur because of low-permeability zeolitic layers and perched water. The model uses the dual-permeability conceptual model, as discussed in Section 7.2.1 and Chapter 2. The following paragraphs summarize the most important features of the mountain-scale flow model for the UZ.

For the base case flow model, the only hydrologic parameters varied are the van Genuchten air-entry parameter (α_f) for fractures (related to fracture aperture) and the fracture-matrix coupling strength (X_{fm}). For each hydrogeologic unit a range of uncertainty was developed for the van Genuchten air-entry parameter based on measurements of fracture density and air permeability. The fracture-matrix coupling strength was not varied independently but was used as a calibration variable. The fracture-matrix coupling strength was constrained to have only three different values (for welded tuff, nonwelded nonzeolitic tuff, and nonwelded zeolitic tuff) in a given calibration. These three values were chosen to optimize the match with observed borehole matrix saturations and water potential. Additional information on parameters and calibration can be found in Chapter 2.

The base case includes five independently calibrated models composed of maximum or minimum air entry parameter for base infiltration divided by three or base infiltration times three, plus a case with the base infiltration and expected value fracture air-entry parameter for each model unit. Five models were used, rather than the full matrix of nine combinations, to reduce computational requirements. Flow was calculated for each of the five models for the three climate states: dry (present day), LTA, and superpluvial. The probabilities for each case are discussed in Chapter 2.

In addition to the base case, an alternative model called the dual-permeability/Weeps model was considered. This model has a significantly reduced fracture-matrix coupling strength to induce more fracture flow. The biggest difference between the base-case model and the dual-permeability/Weeps model is that the dual-permeability/Weeps model shows flow predominantly in fractures even in nonwelded hydrogeologic units while the base case shows flow in nonwelded (nonzeolitic) units predominantly in the rock matrix (see Section 7.6.1.2.4).

We convert TOUGH2 flow fields into a format that can be used by FEHM to perform radionuclide transport calculations using the particle tracking method. The TOUGH2 flow fields provide the node to node fluxes, including fluxes between fracture and matrix nodes, used by FEHM particle tracking. For more details on this interface, see Section 2.1.1.7.

7.5.3.2 Radionuclide Sorption

Sorption is a term used to describe several physicochemical processes including ion exchange, adsorption and chemisorption. Evaluating effectiveness of sorption in performance assessment

for retardation of radionuclide movement through the UZ requires a quantitative, theoretical sorption model. A combination of experimental sorption measurements and modeling has been used to provide this quantitative description for use in performance assessment (Triay et al. 1997; Section IVA).

The main experimental program for sorption was carried out using batch sorption experiments. In these experiments, rock samples were pretreated with groundwater from well J-13, UE-25 p#1, or a synthetic bicarbonate water. These water compositions are expected to overlap much of the range of compositions that may occur in the UZ (Meijer 1992, p. 18; Triay et al. 1997, Section IVD). The pretreated solid phase was then dewatered by centrifugation and saturated with a solution having similar composition, where the compositional difference was the addition of a dissolved radionuclide for sorption measurements. After equilibration, the radionuclide concentration of the solution was measured and the difference between the initial and final concentrations was used to find the amount sorbed (by difference). The experiments may be used to directly evaluate the partition coefficient, K_d, which is defined as the ratio of the molar quantity of radionuclide sorbed per gram of solid phase to the molar quantity dissolved per ml of solution. Experimental uncertainties can be large (Triay et al. 1997, Section IVA p. 99), particularly for small or large values of K_d. Therefore, the values of K_d are commonly reported to one significant figure.

For some radionuclides, specific sorption measurements using rock and water samples from Yucca Mountain have not been performed. For these radionuclides, sorption behavior has been evaluated on a theoretical basis.

Three assumptions were used to develop values of K_d for use in performance assessment (Triay et al. 1997, Section IVD). First, the water compositions from wells J-13 and UE-25 p#1 are assumed to provide an adequate range of compositions to capture the influence of water compositional variability on sorption in the unsaturated. Second, the effects of mineral variations on sorption in the UZ may be approximated using three rock types, devitrified tuff, vitric tuff, and zeolitic tuff. Finally, the effects of temperature are bounded by measurements of sorption at ambient temperature (i.e., that sorption is stronger at elevated temperatures). Elevated temperatures may be expected due to repository heating. However, the effects of mineral alteration due to thermal history are not included in the evaluation of this assumption.

Table 7-3 gives the sorption distribution coefficients recommended for TSPA (Triay et al. 1997, Section IVD). Inspection of the data indicates that we have made no changes to the recommendations provided for TSPA-95, except in the case of Np and Nb in the UZ.

The inherent assumption for the new set of Np sorption values on zeolitic rock is that the carrier water in the UZ will be a dilute water with near-neutral pH (such as J-13). This results in a change in the expected value and maximum value for the K_d of Np on zeolitic rock from 0.5 to 4 and 3 to 12, respectively. The expected value for TSPA-95 was based on sorption in a Paleozoic carbonate water composition (well UE-25 p#1), which resulted in a discrepancy between the relative expected values of sorption coefficients in the devitrified, vitric, and zeolitic units. That is, the minimum value strategy gave values that were lower in the zeolitic unit than in the vitric and devitrified units. However, most of the data and expert judgement indicated that the zeolitic

unit would typically have the highest expected value for K_d. For that reason, the expected value for Np sorption on zeolitic rock was set according to experimental results in J-13 water.

All of the recommendations were provided for oxidizing conditions. If reducing conditions are invoked, the sorption coefficients for Se, Tc, U, and Np would be in the range of 10 to 100 ml/g with an expected value of 20 ml/g for all rock types. The base case will assume oxidizing conditions, which are expected for the UZ environment (including the near-field geochemical environment, see Section 2.5). For Nb, we have conservatively used a K_d of 0 for all rock types because the predominant Nb chemical species for values of pH greater than 7 is expected to be $Nb(OH)^-_6$ (Triay et al. 1997, Section IVA). This negatively charged Nb species is expected to have limited sorption on rock matrix.

Due to limited information available to constrain sorption on fracture surfaces, we conservatively assume that there is no fracture sorption (i.e., the fracture retardation coefficient, $R_f = 1$). Additional information on the effects of fracture sorption on radionuclide transport in the UZ are given in Section 7.6.1.1.3.

7.5.3.3 Matrix Diffusion

The values to be used for matrix diffusion have been measured by Triay et al. (1997). The measurements (in water-saturated rock) found that tritiated water diffused at a rate between 1×10^{-10} m^2/s and 3.5×10^{-10} m^2/s in several samples of devitrified tuff. For Tc, the matrix diffusion coefficient varied between 1×10^{-11} m^2/s and 4.9×10^{-11} m^2/s, which is a lower diffusion rate than for Tritium. The lower diffusion rate is believed to be caused by exclusion from some matrix pores due to the larger ion size and negative charge of the pertechnetate anion (TcO_4^-, the predominant aqueous species of Tc) as compared with Tritium.

The distribution for Tc diffusion coefficients has been assigned to account for variations in rock type and water content. Figure 7-25 summarizes the UFA experimental data (LANL 1997b, pp. VI-5 and VI-6) for conservative tracers and suggests that it is possible to generalize the variation of diffusion with volumetric water content and parameterize it as shown in Figure 7-26. These experiments indicate the magnitude of variation expected as a function of rock type and water content. Water contents of the rock matrix in the UZ generally vary between 0.1 and 0.3. Using the parametric relation for diffusion coefficient as a function of water content gives a range of diffusion coefficients between 3.3×10^{-11} m^2/s to 3.8×10^{-10} m^2/s, or a range of about a factor 10. This range is roughly captured within the 99 percent probability limits (2.5 standard deviations) for a mean value of 3.2×10^{-11} m^2/s and a standard deviation of 1.0×10^{-11} m^2/s. Furthermore, the value of the diffusion coefficient is expected to fall between a minimum value of zero and a maximum value of 10^{-9} m^2/s (which is an approximate value for the diffusion coefficient of dissolved ions in water). We use a beta distribution to capture these aspects of the matrix diffusion coefficient. The diffusion coefficient for other anionic elements, C, Cl, Se, Nb, and I are also assigned values from the distribution for Tc.

Predictions of radionuclide transport for cationic radionuclides (all elements in Table 7-3 except C, Cl, Se, Nb, I, and Tc) using the diffusion coefficient for tritium and measured batch sorption K_ds were found to be conservative relative to measured diffusion behavior, i.e., the predicted rates of uptake in rock matrix were slower than the measured values. The more rapid measured

uptake of sorbing nuclides is believed to be due to deviations from the linear K_d model for sorption. Based on these findings, the diffusion coefficients for sorbing nuclides have been conservatively assigned values representative of Tritium. As described above, the variations in rock type and water content are expected to result in variations in the diffusion coefficient of about one order of magnitude. This range is roughly captured within the 99 percent probability limits (2.5 standard deviations) for a mean of 1.6×10^{-10} m^2/s and a standard deviation of 0.5×10^{-10} m^2/s. This is captured in a beta distribution using the same maximum and minimum constraints as for the anionic elements discussed above.

7.5.3.4 Dispersion

Dispersion coefficients for transport processes in porous media are typically represented by the following relationship:

$$D_i = \alpha_i u_i$$

where D_i is the dispersion coefficient in the ith direction (i.e., in the direction of flow and the two transverse directions) having dimensions of length squared per unit time, α_i is a dispersivity of dimensions length (in the ith direction), and u_i is the flow velocity (in the ith direction) of dimensions length per unit time. Such relationships have been in standard use for transport in a saturated porous medium (e.g., Bear 1972, Chapter 10); however, this type of relationship has also been found to be applicable to unsaturated systems as well (Ma and Selim 1996). In the present case, this dispersion model has been applied to a dual permeability transport model with dispersive transport in both fracture and matrix continua.

There is no data that has been measured at Yucca Mountain that is directly applicable to determining dispersivity in the UZ. However, a value of the 20 m over the approximately 300 m UZ travel distance is consistent with the dispersivity vs. scale correlation of Neuman (1990). The longitudinal dispersivity (primarily in the vertical direction) is not expected to have a large effect on radionuclide transport processes in the UZ because of the strong dispersive effects of fracture matrix interaction that are explicitly captured in the FEHM particle tracking calculation (CRWMS M&O 1997). Also, due to the geometry of the laterally extensive repository source relative to the distance to the water table, the transverse dispersion is not expected to have much influence on radionuclide transport. However, sensitivity analyses are planned in order to provide a firmer basis for these expectations. The base case distribution for dispersivity has been selected to be normal with a mean of 20 m and a standard deviation of 5 m, which captures a range of 7.5 m to 32.5 m within the 99 percent probability limits. This dispersivity distribution applies to transport in all and for both the fracture and matrix continua, but values for fracture and matrix are sampled independently for TSPA calculations.

7.5.3.5 Fracture Aperture

As discussed in Section 7.4.2.3, the model for matrix diffusion of radionuclides between the fracture and matrix continua requires specification of fracture aperture. For a continuous, parallel fracture pattern, the inverse of the fracture aperture is the area of contact between the fracture and matrix continua per unit volume of fracture pore space. Pneumatic fracture

apertures derived by Bodvarrson et al. (1997) mostly lie between about 100 to 200 microns, with some ranging up to 500 microns (Bodvarrson et al. 1997, Table 7.12). However, the pneumatic apertures tend to be dominated by the larger values because flow in fracture networks is proportional to the fracture aperture to the third power (Bodvarrson et al. 1997). Although this may be appropriate for a saturated system (leading to a flux-weighted average fracture aperture), the fractures are typically expected to be unsaturated. Therefore, the fracture apertures through which water is flowing are expected to be somewhat smaller. Therefore, as a first approximation, a lognormal distribution is used with a geometric mean of 1.0×10^{-4} m and a geometric standard deviation of 3.16 (such that a two standard deviation width is one order of magnitude).

7.5.3.6 Bulk Colloid Partition Coefficient

As discussed in Section 7.4.5, Pu is assumed to partition between a dissolved aqueous condition, sorbed to matrix rock, and sorbed to mobile colloids. The bulk colloid partition coefficient, K_c, is computed from the product of the sorption K_d for aqueous Pu onto the colloidal solid times the mass concentration of colloids suspended in solution. The ranges for K_d of Pu on colloids and the colloid mass concentrations are given in Chapter 6. Use of these parameter ranges leads to a range for K_c from 10^{-5} to 10 (dimensionless), and a log-uniform frequency distribution. This distribution is used for both the fracture and matrix continua.

7.5.3.7 Colloid Filtration Coefficient

Due to a lack of information on the filtration process for colloids transporting in the UZ, it is conservatively assumed that there is no filtration in either the fracture or matrix continuum, which is equivalent to a value of $R_c = 1$.

7.6 SENSITIVITY STUDIES

This section investigates various sensitivities and approximations that have been used to establish the TSPA-VA model for UZ radionuclide transport. The sensitivity investigations are intended to give a qualitative understanding of how sensitive the radionuclide mass flux at the water table is found to be with respect to the expected range of variation of parameters associated with the various radionuclide transport processes. The parameters investigated are matrix diffusion, matrix sorption, fracture sorption, and dispersion. The TSPA-VA UZ transport model also uses some approximations to simplify the transport model. An important approximation for the particle tracking method is the linear sorption model for radionuclide interaction with the rock matrix. A sensitivity investigation of this approximation considers the effects of more complex geochemical interactions and different geochemical environments on the transport process. The TSPA-VA UZ radionuclide transport model includes the effects of flow transients caused by climate change using the quasi-steady flow and transport approximation discussed in Section 7.4.4. However, shorter-term or episodic transient flows for a given climate are also expected to be present in the system at some level because rainfall, and hence infiltration, at the surface is not a steady process. A sensitivity study concerning the extent to which the episodic transient flow may influence UZ radionuclide transport as compared with such transport in a steady flow system is investigated. Finally, the use of a minimum K_d model for radionuclide interaction with the rock may suggest that small values of sorption, say on the order of 1 ml/g,

may be negligible and could be assigned a value of zero as a conservative approximation. However, the value of K_d only represents part of the process that determines the importance of sorption on radionuclide transport. Radionuclide contact with the sorptive rock also affects the degree of retardation afforded by sorption. A sensitivity study is conducted to investigate the interplay of radionuclide contact and sorption for a case in which the these factors are believed to be correlated.

7.6.1 Radionuclide Transport Sensitivity Studies

Two factors that affect radionuclide transport in fractured rock media are matrix diffusion and matrix sorption. Previous studies of UZ transport have shown that matrix diffusion and matrix sorption can play a key role in retarding the movement of solutes in fractured porous media (Robinson et al. 1997, Section 9.7). However, the degree of influence of these transport processes on the radionuclide mass flux at the water table is dependent on not only the combination of diffusion and sorption, but also on the source release profile from the potential repository and the UZ flow field (Robinson et al. 1997, Chapter 10). These sensitivity studies are extended here to consider further combinations with FEHM-derived flow fields in Section 7.6.1.1, and to investigate the sensitivity using the TSPA-VA flow fields in Section 7.6.1.2.

Another factor that can also contribute to the performance of the UZ is fracture surface sorption. The limited data from laboratory experiments show that specific coatings on the fracture surface can sorb certain radionuclides and retard the movement of radionuclides in the system.

In our study, we focus on radionuclide transport in unsaturated fractured media. Factors that affect radionuclide transport are matrix diffusion coefficients, radionuclide residence time in fractures, matrix sorption coefficients, fracture surface sorption coefficients, and the relative magnitudes of fracture and matrix flow fractions. In this section, we carry out simulations to study the sensitivity of radionuclide transport to matrix diffusion, matrix sorption, and fracture surface sorption under different flow conditions and different repository release durations.

7.6.1.1 Radionuclide Transport Sensitivity Calculations Using Flow Fields Generated with FEHM

In the sensitivity analysis, we use both 3-D and 2-D models. The flow and transport calculations that we employ in this sensitivity study were computed using the unsaturated flow and transport code FEHM (see Section 7.4 and Robinson et al. 1997). FEHM flow fields were used to allow the investigation of UZ radionuclide transport sensitivities prior to establishing the methods for using TOUGH2 with the FEHM particle tracking method as described in Sections 7.4 and 7.5. Although some differences have been noted between flow fields generated with FEHM versus TOUGH2 (see Chapter 2), we believe that the range of behaviors found between the results for the two codes lies within our range of uncertainty concerning UZ flow. Therefore, transport sensitivity calculations conducted using FEHM-generated flow fields are applicable to TSPA-VA.

We performed the transport calculations in FEHM using the particle tracking technique. The 3-D flow model (grid and hydrogeologic properties) we use here is taken from Robinson et al.

(1997). The flow parameters (Table 15-7, Robinson et al. 1997) are based on one of the parameter set 6.5.4-1 given by Bodvarsson et al. (1997). This parameter set is based on fracture-matrix contact area reduction factors equal to the upstream fracture relative permeabilities in the welded units, but in the nonwelded units (PTn and CHn) the reduction factor is taken to be the upstream (fracture) saturation. We use the Flint infiltration map (Flint et al. 1996) with spatially distributed infiltration rates at the top of the mountain as the upper boundary condition.

In addition to the parameter set discussed above, Bodvarsson et al. (1997) also provided parameter sets using a constant fracture-matrix reduction factors instead of the fracture relative permeability/upstream saturation reduction scheme. We used a 2-D east-west cross section model (northing 232,100 m) to investigate the sensitivity of radionuclide transport to matrix diffusion, adsorption, and fracture surface adsorption under different infiltration rates. The parameter values for the 2-D model is listed in LANL milestone Table 15-3 (Robinson et al. 1997) with fracture-matrix reduction factors from LBNL milestone 6.4.1-2 (Bodvarsson et al. 1997).

7.6.1.1.1 Sensitivity Analysis of Radionuclide Transport to Matrix Diffusion

The strength of matrix diffusion depends on the molecular diffusion coefficients of the radionuclides, the radionuclide resident time in fractures, the fracture spacing, and matrix water contents. Among these factors, the influence of water contents and molecular diffusion coefficient can be represented by the effective matrix diffusion coefficient, which equals the product of molecular diffusion coefficient and water contents. The effect of radionuclide resident time on matrix diffusion is determined by the flow velocity in fractures, which is controlled by the infiltration rates at the top of the mountain.

In the sensitivity study, we fixed the fracture aperture at 10^{-4} m and assumed zero sorption on fracture surfaces or rock matrix. Simulations were run with the effective matrix diffusion coefficient varied from 10^{-11}, 10^{-12}, 10^{-13}, 10^{-14} to 10^{-30} m^2/s to reflect the variations in matrix water content and possible coating effect on the fracture surface. The very low value of 10^{-30} m^2/s is used to represent zero matrix diffusion. Three infiltration maps were used: the Flint base-map infiltration (Flint et al. 1996), 3 times the Flint infiltration, and Flint infiltration divided by 3, to study the effect of flow rate variations on matrix diffusion.

Figures 7-27 and 7-28 show the simulated mass flow rate and the normalized cumulative breakthrough curves at the water table of the 3-D model, respectively, under the Flint infiltration condition (Flint et al. 1996) and a constant radionuclide release at the repository from 0 to 1,000 years. The 3-D model with the fracture relative permeability/upstream saturation reduction factors shows an extremely fast solute transport process in the fractures, which partly contradicts the field observations (see Section 7.6.4). Although we have done sensitivity analyses for the 3-D model, we will focus our analysis on the 2-D results from the East-West cross section model with constant fracture-matrix reduction factors, which were used by LBNL to build the TSPA-VA base-case flow model (see Chapter 2).

Figures 7-29 and 7-30 are the mass flow rate curves and the corresponding normalized accumulative breakthrough curves at the outflow boundary (water table) for the 2-D simulations using the Flint infiltration (Flint et al. 1996) and a release period of 1,000 years. Figure 7-30

shows that around 75 percent of the radionuclides were carried out of the UZ through fractures and the rest were carried through the matrix. This can be seen from the break in slope and the cumulative curve marks the transition from fracture-dominated to matrix dominated transport. It is clear that matrix diffusion slowed down the movement of the radionuclides in fractures and reduced the peak mass flow rate. The strength of matrix diffusion depends on the radionuclide effective diffusion coefficients. For this 2-D flow model with constant fracture-matrix contact area reduction factors, matrix diffusion only begins to play a role in the transport process when the effective diffusion coefficient D_m is greater than 10^{-12} m^2/s. For matrix diffusion coefficients less than this value, the effect of matrix diffusion is negligible. Matrix diffusion also increases the amount of radionuclides transported through matrix as shown in Figure 7-30. The amount of radionuclides moving through the matrix increased from around 25 percent to 40 percent.

The relationships between the matrix diffusion coefficient and the peak mass flow rate and the peak arrival time are plotted in Figures 7-31 and 7-32, respectively. We find that matrix diffusion has little effect on the peak arrival time but can reduce the peak mass flow rates for short-duration releases.

A longer release period reduces the peak mass flow rates and increases the peak arrival time. Figures 7-33 and 7-34 show the mass flow rate curves and the normalized cumulative breakthrough curves for the case with a constant repository release period of 5,000 years. Note that the simulations discussed here for a longer release period are using the same total mass released from the repository. Therefore, we are extending the release period without changing the total mass released. This provides sensitivity of the UZ transport results to the performance of the engineered barrier system. Compared to those with a release period of 1,000 years in Figures 7-29 and 7-30, the peak mass flow rate in this case is reduced from 4.2×10^{-4} mol/year to 1.54×10^{-4} mol/year, and the peak arrival time is increased from 1,200 years to 3,000 years just due to the increase in the release period. The effect of matrix diffusion shows the same pattern as for the 1,000 year release case. For a 10,000 year release period, we find a further decrease in the peak mass flow rate and delays in the peak arrival time, but the effect of matrix diffusion is the same (Figures 7-35 and 7-36).

Figures 7-31 and 7-32 show the relationships between D_m and the peak mass flow rate and the relationship between D_m and the peak arrival time for different release periods. In general, radionuclide transport is not sensitive to variations in matrix diffusion coefficients. Prolonged release periods reduce the peak mass flow rate and delay the peak arriving time, which has little effect on matrix diffusion.

Reducing infiltration rates results in decreasing fracture flow rates, and leads to slower radionuclide transport and reduced peak mass flow rates. Figures 7-37 and 7-38 give the mass flow rate and the normalized cumulative breakthrough curves for the 2-D model with 1/3 of the Flint infiltration rate (Flint et al. 1996) at the top of the mountain. For the 1,000 year release period case, the normalized cumulative breakthrough curves show that the amount of radionuclide carried in fractures is reduced from 75 percent for the Flint infiltration rate to 50 percent. The peak mass flow rate is down from 4.20E-4 mol/year under the Flint infiltration to 1.37E-4 mol/year, and the peak arrival time is increased from 1,200 years to 2,400 years. The second peak in the mass flow rate plots is possibly due to fracture to matrix flow in CHn units. Again, the effect of matrix diffusion is negligible for D_m less than 10^{-12} m^2/sec.

Increasing the release period from 1,000 years to 5,000 and 10,000 years further reduces the peak mass flow rate and delays the peak arriving time at the bottom of the UZ. The effect of matrix diffusion shows the same pattern as those for the cases with the Flint infiltration rate. Figures 7-39 and 7-40 are plots of the relationship between D_m and the peak mass flow rate and between D_m and the peak arriving time, respectively.

Increases in the infiltration rate will increase the amount of water flowing through fractures and accelerate the movement of radionuclides, thus, increasing the peak mass flow rate and reducing the peak mass arrival time. For a radionuclide release period of 1,000 years, the mass flow rate curves and the corresponding normalized cumulative breakthrough curves of different matrix diffusion coefficients for infiltration at three times the Flint base-map infiltration (Flint et al. 1996) are plotted in Figures 7-41 and 7-42, respectively. Compared to the Flint infiltration rate case, the peak mass flow rate is almost doubled from 4.2×10^{-4} mol/year to 8.1×10^{-4} mol/year, while the peak arrival time is basically the same, around 1,000 years. Figure 7-42 shows that nearly 87 percent of the radionuclides are carried in fractures with the remaining 13 percent in matrix. When D_m is greater than 10^{-12} m^2/s, matrix diffusion begins to play a role in reducing the peak mass flow rate and delaying the peak arrival time. For longer radionuclide release periods, reduced peak mass flow rates and delays in the peak arrival time, but these longer release periods have little effect on the sensitivity of transport to matrix diffusion. Figures 7-43 and 7-44 show the relationship between D_m and the peak mass arrival time and between D_m and the peak mass flow rate, respectively, under different radionuclide release conditions.

Our sensitivity studies, based on the 2-D base case flow model, shows that radionuclide transport is not very sensitive to variations in matrix diffusion coefficients. Matrix diffusion only plays a minor role in reducing the peak mass flow rate, and it has little effect on the peak mass arrival time. Increasing the radionuclide release period or decreasing the infiltration rates can greatly reduce the peak mass flow rate and delay the arrival time of the peak mass.

7.6.1.1.2 Sensitivity Analysis of Radionuclide Transport to Matrix Sorption in the Absence of Matrix Diffusion

Matrix sorption can play a significant role in retarding the movement of radionuclides carried in matrix flow. The sensitivity of radionuclide transport to matrix sorption is investigated by fixing $D_m = 10^{-30}$ m^2/s and fracture surface sorption coefficient $K_a = 0$, but by varying the matrix sorption coefficient, K_d, in CHn zeolitic layers from 0 to 100 (31 values) and radionuclide release periods of 1,000 years, 5,000 years, and 10,000 years. Simulations were run using the Flint base-map infiltration rate, 1/3 of the Flint infiltration rate, and three times the Flint infiltration rate (Flint et al. 1996) to reflect the influence of infiltration on radionuclide transport and matrix adsorption.

Under the Flint base-map infiltration condition (Flint et al. 1996) and a release period of 1,000 years, matrix sorption has no effect on the peak mass flow rates and the peak mass arrival times (Figure 7-45). The normalized cumulative breakthrough curves in Figure 7-46 indicate that matrix sorption can greatly retard the movement of radionuclides in matrix but has no effect on radionuclide moving in fractures. When the matrix sorption coefficient was increased from 0 to 2.5, the arrival time of radionuclide in matrix is delayed from 5,000 years to 100,000 years.

Thus, matrix sorption can significantly retard the transport of radionuclides in matrix but has almost no effect on radionuclides in fractures.

Increasing the release period reduces the peak mass flow rate and delays the peak mass arrival time as shown in Figures 7-47 and 7-48. In this case, the release period is increased from 1,000 years to 10,000 years. Note that we are extending the release period without changing the total mass released in this sensitivity calculation. This provides sensitivity of the UZ transport results to the performance of the engineered barrier system. For no matrix sorption, the slow release of radionuclides and fracture to matrix flow results in a delayed high peak mass flow rate at the water table. If present, matrix sorption effectively retards the movement of radionuclides moving from fractures to matrix. Thus, the high peak observed at $K_d = 0$ is eliminated and only see the peak caused by radionuclides moving through fractures can be seen.

The relationships between K_d and the peak mass arrival time and between K_d and the peak mass flow rate under Flint base-map infiltration conditions (Flint et al. 1996) are shown in Figures 7-49 and 7-50, respectively. We find that matrix adsorption alone has no effect on the peak mass flow rate and the peak mass arrival time which is determined by radionuclide transport in fractures. Thus, the peak mass flow rate and the peak mass arrival time are not sensitive to variations in matrix sorption coefficient. Similar behaviors are also observed at 1/3 and 3 times of Flint infiltration rates.

7.6.1.1.3 Sensitivity Analysis of Radionuclide Transport to Fracture Surface Sorption

Laboratory experiments have found that specific coatings on fracture surface can significantly retard the transport of radionuclides in fractures (Triay et al. 1997). Sensitivity studies were carried out to investigate the influence of fracture surface adsorption on radionuclide transport. The fracture surface retardation factors, R_a, in each layer of the CHn units were varied from 0 to 1,000 uniformly. Simulations with fracture surface sorption only were run under different infiltration rates and different radionuclide release periods. In these sensitivity calculations, matrix sorption was set to zero and matrix diffusion was set to 10^{-30} m^2/s.

The simulated mass flow rate curves and the normalized cumulative breakthrough curves for different fracture surface retardation factors under Flint infiltration rate (Flint et al. 1996) and a release period of 1,000 years are shown in Figures 7-51 and 7-52, respectively. Fracture surface sorption has little effect on the peak mass flow rate and the movement of radionuclides in matrix, but can effectively retard the peak mass arrival time.

Increasing the release period can significantly reduce the peak mass flow rate and delay the peak arrival time. Figures 7-53 and 7-54 are the mass flow rate curves and the normalized cumulative breakthrough curves, respectively, for a release period of 10,000 years. In this case, fracture surface sorption plays the same role in retarding the movement of radionuclides in fractures as it does in the 1,000 year release period case. Since the peak mass flow rate in this case is due to fracture to matrix flow, fracture surface sorption has no effect on the peak mass flow rate and the peak arrival time as shown in Figure 7-53.

The relationships between fracture surface retardation factors and the peak arrival time for the Flint base-map infiltration rate (Flint et al. 1996) are shown in Figures 7-55 and 7-56,

respectively. Clearly, fracture surface retardation has little effect on the peak mass flow rate under different radionuclide release period scenarios. For the 1,000 and 5,000 year release conditions, fracture surface sorption can retard the movement of radionuclides in fractures, thus, delaying the arrival time of the peak mass flow rate. But compared to the delay caused by the long release of radionuclides, the delay caused by fracture surface sorption is not significant. For the 10,000 year release period case, fracture surface sorption almost has no effect on the peak arrival time. Similar behavior is also observed for cases with 1/3 of Flint infiltration rate and three times of Flint infiltration rate. From this study, we conclude that for the specific 2-D flow field, radionuclide transport is not very sensitive to variations in fracture surface sorption. However, an important limitation on the sensitivity analysis conducted here is that the range of appropriate retardation factors for fracture surface sorption are basically unknown at this time.

7.6.1.1.4 Sensitivity Analysis of Radionuclide Transport to the Combined Effects of Matrix Diffusion and Matrix Sorption

In order for matrix sorption to play a role in retarding the movement of radionuclides in the geological media, the radionuclides have to be in contact with the matrix. In the fractured media at Yucca Mountain, there are two mechanisms for fracture-matrix interactions: direct fracture/matrix flow exchange and matrix diffusion. Direct fracture/matrix flow exchange carries radionuclides between fractures and matrix by advective transport. In the case where no direct fracture/matrix flow is present, matrix diffusion can play an important role in moving radionuclides from fractures into the matrix driven by the concentration gradient between fractures and the surrounding matrix. These two mechanisms transport radionuclides from the fast fracture flow system into the slow matrix flow system, which can contribute to the performance of the UZ. If matrix sorption is present, the movement of radionuclides in matrix can be further retarded. Since the current base case model is dominated by fracture flow and weak fracture-matrix flow exchanges, the combined effect of matrix diffusion and matrix sorption can be very important in retarding the movement of radionuclides.

We carried out a series sensitivity studies to address the importance of the combined effects of matrix diffusion and matrix sorption. In the simulation runs, fracture surface retardation factors were set to 1, matrix diffusion coefficients were varied from 10^{-30} to 10^{-11} m^2 /s; matrix sorption coefficients in the CHn zeolitic layers were varied from 0.125 to 100; the radionuclide release periods were varied from 1,000 to 10,000 years. Three infiltration rates were used: Flint base-map infiltration rate (Flint et al. 1996), 1/3 of the Flint infiltration rate, and three times the Flint infiltration rate. The influence of different flow conditions on the combined effects of matrix diffusion and matrix sorption were investigated.

We started our analysis with Flint base-map infiltration rate (Flint et al. 1996) and a release period of 1,000 years. Figures 7-57 and 7-58 are the mass flow rate curves and the normalized cumulative breakthrough curves for the case with a matrix sorption coefficient of 0.5 ml/g and different matrix diffusion coefficients. If compared to the case with matrix diffusion only (Figure 7-29) or with matrix sorption only (Figure 7-45), it was found that for matrix diffusion coefficients larger than 10^{-12} m^2/s, the combined effects of matrix diffusion and matrix sorption can reduce the peak mass flow rate. Such an effect is possibly due to the fact that matrix sorption can effectively reduce the amount of radionuclides in the matrix water, and therefore, help maintain a large concentration gradient between the matrix and fractures, which in turn

enhances the effect of matrix diffusion. As a result, the combined effects of matrix diffusion and matrix sorption are more significant than the simple superposition of the effects of matrix diffusion and matrix sorption.

Increasing the matrix sorption coefficient further reduces the peak mass flow rate and delays the peak arrival time. Figures 7-59 and 7-60 give the mass flow rate and the normalized cumulative breakthrough curves, respectively, for a matrix sorption coefficient of 2.5 and a release period of 1,000 years. At K_d =10, the peak mass flow rate is reduced to half of the peak mass flow rate for no matrix diffusion and no matrix sorption conditions (Figures 7-61 and 7-62).

Under the same infiltration condition, increasing the release period does not enhance the combined effects of matrix diffusion and matrix sorption. For K_d =2.5 and a release period of 10,000 years, the peak mass flow rate curves (Figure 7-63) and the normalized cumulative breakthrough curves (Figure 7-64) indicate that the peak mass flow rate is not sensitive to the combined effects of matrix diffusion and matrix sorption.

Since the combined effects of matrix diffusion and matrix sorption are small for matrix diffusion coefficients D_m less than 10^{-12} m^2/s, the only relationships plotted were between the peak mass arrival time and K_d and the peak mass flow rate and K_d at $D_m = 10^{-11}$ m^2/s in Figures 7-65 and 7-66, respectively. It is clear that radionuclide transport is sensitive to the combined effects of matrix diffusion and matrix sorption, only for the fast release condition (for a release period of 1,000 years). When the matrix sorption coefficient is larger than 20, the radionuclide transport process is less sensitive to the combined effects. For a release period larger than 5,000 years, the combined effects have little influence on the peak mass flow rate or the peak mass arrival time.

A decrease in infiltration rate, decreases the amount of flow in the fractures. This results in increased radionuclide resident times in fractures, and hence increases the strength of fracture-matrix interactions due to matrix diffusion and matrix sorption.

Figures 7-67 and 7-68 are the mass flow rate and the normalized cumulative breakthrough curves using 1/3 of the Flint infiltration rate (Flint et al. 1996) and a release period of 1,000 years. For K_d =2.5 and D_m =10^{-11} m^2/s, the peak mass flow rate was reduced almost to half of the value under the same flow condition, but without matrix sorption or matrix diffusion. The reduction magnitude is much larger than that under the Flint infiltration condition. For the matrix diffusion coefficient of 10^{-11} m^2/s, increases in the matrix sorption coefficient not only reduce the peak mass flow rate but also result in significant delays in the peak mass arrival time (Figures 7-69 and 7-70 for K_d = 10.0).

The relationships between the peak mass arrival time and K_d and the peak mass flow rate and K_d for $D_m = 10^{-11}$ m^2/s are plotted in Figures 7-71 and 7-72, respectively. In contrast to those for the Flint base-map infiltration case (Flint et al. 1996), radionuclide transport is more sensitive to the combined effects under this lower infiltration rate condition, even for the longer release period of 5,000 years. Similarly, for the 10,000 year release period case and Flint infiltration condition, radionuclide transport under this long release condition is not sensitive to the combined effects.

An increase in the infiltration rate results in increases in the amount of fracture flow and the fracture flow rate, which reduces the radionuclide resident time in fractures. Under three times

of Flint infiltration rate (Flint et al. 1996) and a release period of 1,000 years, the relative reductions in peak mass flow rate and the peak mass arrival time (Figures 7-73 and 7-74) are less than those for 1/3 Flint infiltration rate.

Figures 7-75 and 7-76 present the mass flow rates and the normalized cumulative breakthrough curves for a release period of 10,000 years. Again, increases in the release period do not have significant impact on the combined effects of matrix diffusion and matrix sorption.

The relationships between the peak mass arrival time and the matrix sorption coefficient (Figure 7-77) and between the peak mass flow rate and the matrix sorption coefficient (Figure 7-78) for $D_m = 10^{-11}$ m^2/s indicate that at higher infiltration rates the peak mass flow rate and the corresponding arrival time is most sensitive to the combined effects under the 1,000 year release condition. For release periods larger than 5,000 years, the combined effects have little effect on the peak mass flow rates and the peak mass arrival time.

In general, our study indicate that for a matrix diffusion coefficients larger than 10^{-12} m^2/s and a release period less than 5,000 years, radionuclide transport in the 2-D flow field is sensitive to the combined effects of matrix diffusion and matrix sorption. If the release period is longer than 5,000 years, then the combined effects of matrix diffusion and matrix sorption have little effect on transport. For lower infiltration rates, the corresponding lower-fracture, flow rates, which can enhance the combined effects of matrix diffusion and matrix sorption, reduce the peak mass flow rates and delay the corresponding peak arrival time.

7.6.1.1.5 Influence of Different Fracture-Matrix Flow Configuration on the Sensitivity Studies

The previous sections describe the influence of transport parameters on the radionuclide movement in the specific 2-D flow field. The flow configurations between fractures and the matrix can play a significant role in the transport process of radionuclides. Specifically, we want to understand how different flow configurations are going to affect the strength of matrix diffusion and matrix adsorption. This section illustrates simulations with different fracture and matrix flow configurations that address the influence of flow configurations on matrix diffusion and matrix sorption.

The LANL Strong Fracture-Matrix Coupling Model

To illustrate the importance of flow configuration on radionuclide transport, the Robinson et al. (1995) parameter set (set 2) was used in a similar 2-D model. Compared to the LBNL parameter set (set 1), set 2 has a slightly lower fracture permeability (6.0×10^{-10} m^2 vs. 2.0×10^{-9} m^2 for set 1) in CHn and relative lower matrix permeabilities in CHn zeolitic layers (7.0×10^{-18} m^2 vs. 9.12×10^{-18} to 5.75×10^{-17} m^2 for set 1), higher fracture van Genuchten n parameters for all layers (3 vs. 1.97 for set 1), and no fracture-matrix contact area reduction factors. Application of set 2 parameters in the radionuclide transport simulations resulted in matrix-dominated flow configurations in the Calico Hills unit. This difference in flow configurations resulted in substantially different effects of matrix diffusion and matrix sorption on radionuclide transport.

The Influence of Different Flow Configuration on Radionuclide Transport

Four cases were investigated: no matrix diffusion or matrix adsorption, matrix diffusion ($D_m = 10^{-11}$ m^2/s) but no matrix adsorption, matrix adsorption (K_d =2.5 in the Calico Hills zeolitic layers) but no matrix diffusion, and finally matrix diffusion and adsorption ($D_m = 10^{-11}$ m^2/s and K_d =2.5 in the Calico Hills zeolitic layers). The radionuclides were released uniformly over a period of 5,000 years. All four cases were run using parameter sets 1 and 2, respectively.

The mass flow rate curves for the two parameter sets under different conditions are plotted in Figures 7-79 and 7-80, respectively. Figure 7-79 shows that for fracture dominated flow, the radionuclides travel at faster speed and arrive at the water table at higher concentrations than those in Figure 7-80. This is due to the short residence time of radionuclides in fractures and less fracture to matrix flow. The effect of matrix diffusion and matrix sorption on radionuclide transport is minimal. Matrix sorption alone has no effect on radionuclides travelling in fractures. Figure 7-80 shows how matrix dominated flow and strong flow exchanges between fractures and matrix affect radionuclide transport processes. As for the case in Figure 7-79, matrix diffusion only plays a minor role in reducing the amount of radionuclides carried in fractures, and matrix sorption alone has no effect on radionuclide transport through fractures but, in combination, can greatly reduce the peak radionuclide mass flow rate at the water table and retard the travel time of radionuclides moving through matrix. Compared to the mass flow rate curves from set 1, the combined effects of matrix diffusion and matrix sorption in this case is much more prominent, which is reflected by the large reduction in the peak mass flow rate and the retardation of the peak arrival time. Such strong effects of matrix diffusion and matrix sorption is determined by the relatively low fractional fracture flow in the system. Figure 7-79 and 7-80 both show that the effect of matrix diffusion is reversible. Matrix diffusion can move the sorbed radionuclides back into the fractures, so at some time following the fracture mass flow rate peak, the mass flow rate curves from the combined effects are found to be higher than those for matrix sorption only.

The simulation results from our study indicate that in unsaturated fractured media with fracture dominated flow and relative long radionuclide release periods (5,000 years), matrix diffusion and matrix sorption only have limited effects on radionuclide transport in the system. On the other hand, in a system dominated by matrix flow, matrix diffusion and matrix sorption can significantly reduce the peak mass flow rate and retard the movement of radionuclides. When matrix sorption is present, matrix diffusion plays an important role in enhancing fracture-matrix interactions and retarding the radionuclide transport process in fractures.

7.6.1.1.6 Interpretations

Our sensitivity study shows that for the specific 2-D flow model generated using the FEHM flow code and preliminary LBNL hydrogeologic parameter sets, radionuclide transport in the UZ is found to have low sensitivity to variations in matrix diffusion coefficients, fracture surface sorption, and matrix sorption individually. But the combined effects of matrix diffusion (for $D_m > 10^{-12}$ m^2/s) and matrix sorption can significantly reduce the peak mass flow rates and delay the corresponding peak arrival time. Under the condition of $D_m = 10^{-11}$ m^2/s, radionuclide transport is most sensitive to variations in matrix sorption coefficients in the range of 0 to 20 ml/g.

Increasing the radionuclide release period can effectively reduce the peak mass flow rate and delay the corresponding peak mass arrival time. Such effects are far larger than the effects of matrix diffusion, matrix sorption, and fracture surface sorption on the radionuclide transport

process. Finally, different flow configurations can have a strong impact on the transport of radionuclides in the UZ.

7.6.1.2 Radionuclide Transport Sensitivity Calculations Using TSPA-VA Base-Case and Alternate-Case Flow Fields Generated with TOUGH2

In this section, we investigate the sensitivity of UZ radionuclide transport using the TSPA-VA flow fields generated with TOUGH2. All of these sensitivity calculations are based on a 3-D, dual-permeability flow and transport model, as described in Chapter 2 and Section 7.5.3.1.

7.6.1.2.1 Sensitivity Calculations for Technetium Transport

Sensitivity calculations have been performed to investigate the effects of climate (infiltration) and matrix diffusion on the transport characteristics of technetium (Tc) in the UZ. Here we have used a source term with a duration of 400,000 years, as a representative time scale for releases from the potential repository as it is currently modeled (see Chapter 6). The releases are assumed to be uniform over this time period, and are also uniformly distributed spatially throughout the grid blocks that represent the potential repository. The calculations presented here were performed using 1,000,000 particles and do not include radioactive decay. Figures 7-81 and 7-82 show the behavior of Tc releases at the water table in terms of mass flux and accumulative recovery, respectively. The infiltration is varied according to the climate and infiltration model (see Chapter 2). The average infiltration over the potential repository block for the three climates varies from 7 mm/yr for present day (PD) climate, 42 mm/yr for the long-term average (LTA) climate, and 110 mm/yr for the super-pluvial (SP) climate. The "climate" is fixed for the duration of each realization. Matrix diffusion is varied between 10^{-30} m^2/s to 3.2×10^{-11} m^2/s. The value of 10^{-30} m^2/s was used as a nominal value for zero matrix diffusion. The mass flux response in Figure 7-81 and the accumulative plot in Figure 7-82 show that there is not much sensitivity to infiltration or matrix diffusion. The reason for the lack of sensitivity is that the transport process is relatively rapid under all the combinations of conditions. Therefore, any changes on the time scale represented by the 1 million year plots are nearly undetectable. Note that the more irregular patterns that develop at early times in Figure 7-81 are a result of smaller time steps used to capture the initial breakthrough. As the time steps are increased at later times, the profiles become much smoother. The high-frequency variability at small times seen in the mass flux plots are seen to have little effect on the accumulative recovery profiles in Figure 7-82.

7.6.1.2.2 Sensitivity Calculations for Neptunium Transport

Neptunium (Np) introduces another element of complexity to the transport process through its sorptive interaction with rock matrix surfaces. The simulations for Np are carried out in a manner similar to that describe for Tc. The first set of comparisons shown in Figures 7-83 and 7-84 are for the effects of climate, (i.e., infiltration). As compared with Tc, Np transport shows a much greater sensitivity to changes in infiltration. This is due to the longer transport times for Np and the greater sensitivity of transport time to fracture/matrix interaction. The principle difference between Tc and Np transport is sorption, and it is sorption that makes Np transport significantly slower than for Tc. In these calculations, Np is assigned the base-case average sorption values of 1 ml/g in the devitrified rock, 1 ml/g in the vitric rock, and 4 ml/g in the zeolitic rock. The matrix diffusion values for Tc and Np are 3.2×10^{-11} m^2/s and 1.6×10^{-10} m^2/s,

respectively. Figures 7-85 and 7-86 show the effects of sorption (K_d) and matrix diffusion (D_m) on Np transport under long-term-average climate. (Np sorption coefficients are given in Section 7.5.3.2.) These figures demonstrate that transport is sensitive to matrix diffusion only when combined with sorption. The effects of matrix diffusion without sorption on transport is negligible, as was also found for Tc transport. Greater sensitivity to matrix diffusion is apparent as K_d is increased. The changes in response with increasing sorption are reduced peak mass flux and delayed arrival at the water table.

7.6.1.2.3 Sensitivity Calculations for Plutonium Transport

Plutonium (Pu) is modeled as an aqueous species that can sorb onto either rock surfaces or suspended colloidal material. A model for transport that includes reversible sorption with suspended colloidal material is presented in Section 7.4.5. The simulations were conducted, as for Tc and Np, using a spatially and temporally uniform source of 1,000,000 particles distributed over the potential repository for a duration of 400,000 years. In these calculations, Pu is assigned the base-case average sorption values of 100 ml/g in the devitrified rock, vitric rock, and zeolitic rock. The matrix diffusion values for Pu is 1.6×10^{-10} m^2/s. Figure 7-87 shows the response of Pu to different UZ flow fields that result under different climates. The higher infiltration rates during LTA and SP climates result in significantly shorter travel times and higher peak-mass flux rates at the water table. Very little Pu reaches the water table under the PD climate over 1,000,000 years, as seen in Figure 7-88. In contrast, more than 70 percent of the total released Pu arrives at the water table in the 1,000,000 time period. Note that reversible sorption interaction with colloids is included in these calculations using the expected value (0.01) of the reversible colloid partitioning coefficient, K_c. The effects of sorption (K_d) and matrix diffusion (D_m) are shown in Figures 7-89 and 7-90 for long-term-average climate. Matrix diffusion is found to play a more important role, in general, for Pu than for Tc or Np. Even for the minimum values of K_d, including matrix diffusion is found to reduce the peak mass flux and increase the travel time to the water table. Note that the mass flux curves for the condition of very low (essentially zero) matrix diffusion (10^{-30} m^2/s) show increasing mass flux up until the source term is exhausted at 400 ky. These curves then show a very rapid decline, followed by a gentler increase in the peak mass flux from 400 ky to 600 ky. Finally, the sensitivity to the colloid partitioning parameter, K_c is displayed in Figures 7-91 and 7-92 for LTA climate. The value of K_c ranges from 0 (no colloidal radionuclide transport) to a maximum value of 10, in which 90 percent of the Pu are attached to colloids. As can be seen in the figures, the effects of K_c are most pronounced as K_c rises above the expected value of 0.01 to the maximum value of 10. For values of K_c smaller than 0.01, colloids are found to have much less effect on the transport characteristics of Pu.

7.6.1.2.4 The Effects of Unsaturated Zone Flow Conceptual Model

Calculations were performed to see what influence the UZ flow field conceptual model may have on radionuclide transport. The two flow field conceptual models are the base-case (mean-alpha) flow model and the DKM Weeps (mean-permeability) flow model. A more complete description of these flow models is given in Chapter 2, however, the DKM Weeps flow model results in the greatest flow and transport through the fractures than the base-case flow model. Again the spatially uniform source is used over 400 ky. The results are shown in Figures 7-93 and 7-94. The different flow fields appear to have very little influence on Tc transport. For the time

resolution on these figures, Tc transport for either case is relatively fast, and; therefore, the transport results have only a weak sensitivity to the changes in the flow field. As compared with Tc, somewhat greater sensitivity is found for Np and Pu. These greater sensitivities for transport of Np and Pu to the UZ conceptual flow model are a result of the longer travel times from the potential repository to the water table for these two radionuclides. However, the overall effect of the different flow conceptual models on transport is not large.

7.6.1.2.5 Sensitivity to Rock Type

Differences in hydrogeologic and thermogeochemical properties between the devitrified, vitric, and zeolitic rock types suggest that they may not affect transport in the same way. For this simulation, one mole of Np is released uniformly over the repository region for a period of 10,000 years. As before, the calculations were performed using the mean flow field for the base case and mean infiltration for the LTA climate. The K_ds for Np sorption on the different rock types are 1 ml/g for devitrified rock, 1 ml/g for vitric rock, and 4 ml/g for zeolitic rock. Three transport calculations were conducted with sorption "turned on" in one layer only, that is, K_ds were equal to zero in the other layers, to see the influence of each individual rock type. A final calculation was done using all the base-case K_ds "turned on." The results are shown in Figures 7-95 and 7-96 for the radionuclide mass flux and cumulative mass arrival, respectively, at the water table.

The conclusion based on this sensitivity calculation is that the TSw unit (with sorption parameter dK_d, where the leading "*d*" stands for the devitrified rock type) is the most important unit in delaying radionuclide arrival at the water table. Flow through the TSw unit is nearly vertical throughout the repository domain, and this flow pattern causes longer residence times during transport through the fractures, allowing greater interaction with the matrix. On the other hand, large portions of the Calico Hills unit are bypassed because of lateral diversion. Therefore, in many areas the flux that does pass through the Calico Hills is relatively focused and residence times in these fracture pathways are short, leading to poor interaction with the matrix. The Calico Hills vitric unit generally has the next largest effect on radionuclide retardation because of high matrix permeability that attenuates fracture flow and causes greater radionuclide contact with the matrix.

7.6.1.2.6 Effects of Dispersion on Radionuclide Transport

Sensitivity calculations concerning the effects of dispersion were carried out for the base-case, expected-value UZ hydrogeologic parameter set and the base-case infiltration map for LTA infiltration. The transport parameters used are as follows: no matrix diffusion, no matrix sorption, no fracture sorption, the expected value fracture aperture and no colloids. The values of dispersivity investigated ranged from zero to 75 m. As described in Section 7.4.2.2, the dispersion model for the particle tracking method requires isotropic dispersion. Therefore, the longitudinal and transverse dispersivities are the same. Our current level of understanding is not sufficiently refined to distinguish different levels of dispersion between the fracture and matrix continua or for different hydrogeologic units; therefore, we have used a single value to represent the dispersion process for each simulation. A total of 500,000 particles were used to represent a pulse release of one mole of radionuclides released uniformly throughout the repository grids.

The results show that the effect of dispersion on radionuclide transport is negligible (see Figures 7-97 and 7-98). The normalized accumulative breakthrough curves show a very small effect at small times and virtually no effect at larger times. The limited effects of dispersion on the radionuclide transport process is due to several factors that tend to mask the effects of dispersion in the model. These include variations in stratigraphy, fracture/matrix interaction, and the relatively large, spatially uniform source term. These factors, which are explicitly accounted for through the model, act to disperse the radionuclides and reduce the impact of a dispersion term in the transport model equation.

7.6.1.2.7 Travel Time and Pathways Spatial Patterns in the Unsaturated Zone

Simulations using the UZ transport model were performed to investigate the travel times and transport pathways from the various repository regions to the water table. The simulations used 100,000 particles released from each node in the repository to develop a travel time distribution for transport to the water table. The simulations were also used to identify the center of mass at the water table for releases from each repository node. This information indicates transport pathways through the UZ. The results presented here are for expected infiltration under LTA climate, which averages about 40 mm/year over the repository block. The LTA climate is expected during about 90 percent of the postclosure time period.

Figures 7-99a and 7-99b show the contour plots of the repository region; color represents travel time to the water table. These calculations were done using hypothetical non-sorbing, non-diffusing particles to focus on the advective transport processes. Two plots are presented. Figure 7-99a shows the times for 0.1 percent of the released mass to arrive at the water table as a function of location within the repository. Figure 7-100b shows the times for 50 percent of the released mass to arrive at the water table. The general trend is for the travel time to increase from northeast to southwest through the repository. The shorter travel times in the northeast (less than 50 years for 0.1 percent; less than 200 years for 50 percent) are attributed to transport dominated by the fracture continuum. The northeast section is a zone in which site characterization studies have commonly found perched water along the basal vitrophyre of the TSw unit and upper zeolitic units in the CHn unit. The flow model has been calibrated to reproduce observations of perched water. This calibration was achieved by severely reducing fracture permeabilities in zones immediately underlying zones of observed perched water. This reduction in fracture permeability leads to lateral diversion of the flow through more permeable fracture pathways around the perched water. Further east, the diverted water eventually returns to flow pathways that are mainly vertical, either along the Ghost Dance fault or other fracture pathways. Another factor influencing travel times is the uneven distribution of infiltration over the repository block. Infiltration models predict higher infiltration rates in the northeast than in the southwest (see Chapter 2).

Figures 7-100a and 7-100b are similar plots to Figures 7-99a and 7-99b, but in this case the travel times are for particles having the transport characteristics of plutonium. In the base case, plutonium is assumed to interact with colloids both reversibly and irreversibly. Irreversible sorptive interaction with colloids causes particles to act as nonsorbing and nondiffusing, resulting in travel times similar to those shown in Figure 7-99a and 7-99b. Reversible sorptive interaction with colloids causes different magnitudes and patterns for travel times. Similar qualitative trends in travel time are shown in Figure 7-100a as in Figure 7-99a (both for 0.1

percent recovered mass). However, the magnitudes of the travel times are much larger for the plutonium case. Travel times approach 100,000 years for plutonium, compared with maximum travel times that are less than 200 years for the passive tracer. Travel times for 50 percent of the released plutonium are in excess of 100,000 years, and some exceed one million years. Also, the spatial trends in travel time have changed and are much less distinct.

Figure 7-101a shows the origin points of particle releases from the repository. These points have been color coded so that releases from the six zones (cc ≡ central central; ne ≡ northeast; nw ≡ northwest; sc ≡ south central; se ≡ southeast; sw ≡ southwest) can be identified at the water table in Figure 7-101b. The colored dots in Figure 7-101b represent the center of mass of each of the releases upon arrival at the water table. Lateral diversion of releases from the northern portions of the repository is the most prominent feature. Some diversion is also seen from the southeast portion of the repository. The somewhat abrupt termination of lateral diversion eastward is a manifestation of the end of the perched water body upon which lateral diversion occurs. The linear pattern of releases at the water table originating from the northwest portion of the repository (black dots) may result from the dip of the hydrogeologic units intercepting the water table or, alternatively, a high-permeability feature is interrupting lateral diversion. Transport pathways from the southwest, south central, and central-central regions are more nearly vertical.

7.6.2 Sensitivity Study on the Assumptions of Isothermal Conditions and Linear, Equilibrium Sorption for Radionuclide Transport; an Investigation of ^{237}Np

(Note: much of this section can be found in Robinson et al. 1997, Chapter 11). In addition to the hydrologic processes that are important in controlling radionuclide migration, transport processes such as advection and dispersion, along with geochemical processes such as precipitation-dissolution, adsorption, and aqueous speciation reactions, will also affect the movement of radionuclides. For all but the most soluble radionuclides, the solubility determines the amount of radionuclide that will be mobilized after a canister breach. Many radionuclides participate in aqueous speciation reactions, resulting in the formation of numerous aqueous complexes. These complexes often differ in their adsorption behavior. The interaction among these various geochemical processes may play an important role in radionuclide migration. In general, a numerical model must account for these hydrological, transport and geochemical processes in order to be credible.

This section investigates the migration of ^{237}Np from the potential repository to the water table. Because of its long half life (2.14×10^6 years), relatively high solubility under oxidizing conditions, low dose limits, and predicted low sorption, performance assessment models have identified ^{237}Np as a radionuclide of concern for the proposed repository at Yucca Mountain (Wilson et al. 1994; CRWMS M&O 1995a) and is one of the nine radionuclides addressed in the base-case for TSPA-VA. We use FEHM (Zyvoloski et al. 1992) to study ^{237}Np transport. For this analysis FEHM is used to perform both the thermal-hydrologic calculations and the reactive transport. Because chemical reactions are involved, the differential material balance method (see Section 7.4.1.1) is used for computing transport, as opposed to particle tracking. This allows for simultaneous solution of nonlinear chemical reactions with transport, which is difficult to accomplish with the particle tracking formulation. The reactive transport model for FEHM simulates the important geochemical processes that affect ^{237}Np migration. An equilibrium

formulation is used for aqueous speciation reactions, because the time scale of such reactions are typically much shorter than groundwater transport time scales, making the local equilibrium assumption valid (e.g., Steefel and Lasaga 1994). By contrast, rock-water interactions such as adsorption and precipitation-dissolution reactions often exhibit kinetic limitations in groundwater systems (e.g., Steefel and Lasaga 1994). Therefore, we use a kinetic formulation for these rock-water reactions.

Previous studies (Birdsell et al. 1994; Robinson et al. 1995) have modeled the transport of ^{237}Np using linear equilibrium adsorption models that may have oversimplified these geochemical processes. The transport calculations of this chapter examine the migration of ^{237}Np as a function of groundwater chemistry and composition. Batch and column sorption experiments (Triay et al. 1996a, b, c; Tait et al. 1996) along with speciation and solubility experiments (Wolery 1992; Nitsche et al. 1993; Bertetti et al. 1996; Efurd et al. 1996) are used to construct the reactive transport model. The model is then applied to the 2-D site-scale simulations.

The flow system is in part constrained by recent published data (Flint et al. 1996; Paces et al. 1996; Triay et al. 1996a; Robinson et al. 1996) used to estimate infiltration rate and bound the percolation flux through the UZ. Furthermore, we extend previous transport model predictions to non-isothermal conditions brought on by radioactive decay waste heat. Several studies have addressed the impact of heat on the thermal-hydrologic conditions in the near field (Buscheck et al. 1996) and at the site scale (Buscheck and Nitao 1993) for the purpose of establishing the conditions to be experienced by waste packages. These packages are being designed to withstand a potentially corrosive environment for up to 10,000 years. The site-scale thermal-hydrologic calculations of the present study are designed to study the possible impacts on ^{237}Np migration after waste canisters are breached and radionuclides escape the near-field environment.

The overall goal of this section is to examine the combined effects of infiltration rate, repository heat, and reactive transport processes on the migration of ^{237}Np from the potential repository to the water table. Including these effects allows assessment of the validity of assumptions embodied in the TSPA-VA concerning linear equilibrium sorption and the influence of repository heating on radionuclide transport in the UZ.

Flow and Energy Formulations - Detailed derivations of the governing equations for two-phase flow including heat transfer have been described by several investigators (e.g., Brownell et al. 1975; Mercer and Faust 1975; Zyvoloski 1983). The analyses we present here use FEHM, a multi-dimensional finite element code suitable for simulating systems with complex geometries that arise when modeling subsurface flow and transport. For the 2-D simulations in this section, the relative permeability curves for the fractured rock are assumed to be adequately represented by an ECM. The primary assumption in the model is that the capillary pressure in the fractured medium equals that in the matrix. Assuming the fractured and matrix domains can each be described using the characteristic curve model introduced by van Genuchten (1980), the composite permeability of the medium at a given matrix saturation (and capillary pressure) can be represented as a weighted average of the permeabilities of the two media. From this treatment, a single value can be obtained for the relative permeability. Because fractures are predicted to be dry until the capillary pressure in the matrix reaches a low value (at high saturation), the permeability is dominated by matrix properties until almost complete saturation

of the matrix, after which fracture saturations and liquid permeabilities rise due to fracture flow. Limits of applicability of the ECM model were explored for fluid flow by Pruess et al. (1990).

For transport calculations, these assumptions are less likely to be applicable than for the case of fluid flow because of the longer time scales of solute transport relative to transmission of a fluid pressure difference within a matrix block (Robinson et al. 1997, Chapters 4 and 5). Sensitivities to the dynamic fracture/matrix interaction for transport have been considered using a 1-D, dual permeability model (DKM) (Robinson et al. 1997, Section 11.8.1). Results from the DKM model show that the breakthrough curve is bimodal (due to significant components of both fracture and matrix transport) for percolation flux values of 4 mm/yr. At lower percolation flux rates, the transport in the matrix was found to be dominant. The bimodal breakthrough behavior cannot be reproduced with an ECM model, however a bound for this behavior may be achieved in the ECM model by assuming the zeolite porosity is the same as the fracture porosity. The ECM model is used for the multi-dimensional calculations presented here primarily because of the extreme computational burden imposed by the DKM model when thermal and chemical processes are included.

Reactive Transport Equations - The equations in the reactive transport model are coupled to the heat and flow system because the flow and energy equations are used to obtain flow velocities and temperature-dependent transport properties. However, we assume that transport has a negligible effect on fluid properties, permeability, and the heat balance and do consider possible effects on rock properties due to the mineralogic alteration of rocks and the redistribution of dissolved minerals. Furthermore, the alteration and recrystallization of minerals to phases that are either more sorptive or less sorptive is not addressed here.

FEHM uses aqueous, immobile, and vapor components as the primary dependent variables (PDVs) in the reactive transport equations. A set of chemical components is defined as the minimum number of species that uniquely describe the chemical system (Mangold and Tsang, 1991). In FEHM, the secondary dependent variables (SDVs) are uncomplexed aqueous component concentrations and aqueous complex concentrations. We assume that all aqueous phase speciation reactions are at equilibrium (known as the local equilibrium assumption), resulting in the following relationship between PDVs and SDVs:

$$C_j = c_j + \sum_{i=1}^{N_x} a_{ij}\, x_i \qquad (7\text{-}34)$$

where C_j is the total aqueous concentration of component j, c_j is the uncomplexed concentration of component j, x_i is the concentration of complex i, a_{ij} is the stoichiometric coefficient representing the number of moles of component j in complex i, N_c is the number of aqueous components and N_x is the number of aqueous complexes (Yeh and Tripathi 1989).

Applying the local equilibrium assumption to aqueous-phase speciation reactions greatly reduces the number of PDVs in the transport equations and is generally accepted to be a valid assumption (e.g., Steefel and Lasaga 1994). Given the total aqueous concentration of each aqueous component, the SDVs can be calculated using a set of nonlinear algebraic equations derived from chemical equilibrium theory (e.g., Lichtner 1996). To obtain the total aqueous concentrations,

the reactive transport equations for aqueous, immobile, and vapor components must be solved. There are no SDVs for either vapor or immobile components, because the model as currently formulated treats only aqueous speciation. Reactions involving vapor and immobile components are treated with a kinetic formulation, with kinetic reactions represented as reaction source-sink terms in the reactive transport equations.

Further details concerning the flow and chemical reaction mathematical formulation and solution methods are given in (Robinson et al. 1997, Chapter 11).

7.6.2.1 Minimum K_d Model Abstraction for Radionuclide-Rock Interactions

The sorption coefficient K_d for ^{237}Np measured on core samples in the laboratory varies from effectively 0 to values of order 5 ml/g, over the range of geochemical conditions found in Yucca Mountain fluids and rocks. This range has sufficient impact on performance to motivate the development of a reactive transport model that predicts the adsorption of ^{237}Np as a function of water composition. Considerable experimental work has been done to characterize ^{237}Np solubility, speciation and sorption over the range of conditions expected at the Yucca Mountain site (e.g., Nitsche et al. 1993, Bertetti et al. 1996, Efurd et al. 1996, Triay et al. 1996a). These experimental results have been combined with studies of the Yucca Mountain groundwater and pore-water geochemistry (Ogard and Kerrisk 1984, Triay et al. 1996a, Yang et al. 1996) to develop a reactive transport model for ^{237}Np that includes dissolution, speciation and sorption of ^{237}Np as a function of groundwater composition.

The experimental program for studying ^{237}Np sorption, solubility, and speciation was designed under the assumption that fluids from the SZ in wells J-13 and UE-25 p#1 (Table 7-4) can be used to bracket the composition of the Yucca Mountain fluids in the UZ (Meijer 1992). J-13 water is a dilute sodium bicarbonate water from the shallow SZ. Sodium, calcium, and magnesium are the major cations, and bicarbonate, sulfate, chloride and fluoride are the major anions, in order of decreasing concentration. Silica is the only other major constituent (Ogard and Kerrisk 1984). UE-25 p#1 water is obtained from the SZ from the deeper Paleozoic carbonate aquifer and has higher concentrations of most of these chemical species.

7.6.2.2 Neptunium Chemistry: Solubility, Speciation, and Sorption Models

Solubility - Solubility establishes an upper limit for the concentration of aqueous Np in the source term for radionuclide migration from the near-field environment. Nitsche et al. (1993) and Efurd et al. (1996) measured the solubility of ^{237}Np from groundwaters representative of the Yucca Mountain region. The more recent measurements of Efurd et al. (1996) are considered to be more representative since they employed refined experimental techniques for adjusting solution pH. Solubilities for J-13 groundwaters are shown in Table 7-5.

We model the precipitation-dissolution of Np at the repository with a first order kinetic expression (e.g., Lasaga 1984). The solubility product data at 25, 60, and 90° C are used by the model to estimate the value of the solubility product as a function of temperature using polynomial interpolation. Neptunium dissolution has proven to be fast in comparison to the transport time scales associated with Yucca Mountain (Nitsche et al. 1993). Therefore, we select the kinetic parameter value to ensure that the reaction proceeds to equilibrium.

The solid component represents the total amount of ^{237}Np present at a given repository node and is the source term for any aqueous Np released into the system. The Np inventory for the simulations was computed assuming the initial quantities of individual radionuclides given by Wilson et al. (1994) assuming a 70,000 metric tons uranium (MTU) total inventory. To calculate the mass of Np at each repository node in the 2-D cross section, the total inventory is divided equally into each drift node and divided by the thickness of the repository in the unmodeled direction to place the calculations on a one-meter-thick basis.

In the transport simulations, it is assumed that once the drift node rewets, the canisters immediately are breached and radionuclides escape the near-field environment at the solubility limit of ^{237}Np. Therefore, for simplicity and in the interest of conservatism, it is assumed no retardation effect of the near-field engineered barrier once water contacts the waste packages. If this assumption proves to be overly conservative, similar calculations could be performed, in which the dissolution reactions are initiated after an additional time lag to simulate the performance benefits of the engineered system.

Speciation - Neptunium released into the aqueous phase participates in speciation reactions. These speciation reactions are given in Table 7-6. The three total aqueous components chosen to model these reactions are NpO_2^+, HCO_3^- ,and H^+.

To demonstrate chemical speciation behavior of the system, a series of one-element, batch FEHM simulations were performed. Since there is no flow or transport in these simulations, the results are similar to those of a batch geochemical code such as EQ3/6 (Wolery 1992). Calculations were performed at 5×10^{-6} mol/l of Np, a typical value of solubility. The total carbonate concentration in solution was taken to be either that of J-13 or UE-25 p#1 fluid. Figure 7-102 shows the speciation results for Np in J-13 water at 25° C. At low pH values, the dominant species is NpO_2^+ , with carbonate and hydroxy complexes becoming significant at pH values of about 8 and higher. In the higher carbonate UE25 p#1 water, the results are similar, but the transition to the carbonate and hydroxy complexes occurs at pH values closer to 7. Also, the specific species that are important at higher pH values tend to be the carbonate rather than the hydroxy complexes. As will be demonstrated below, the fraction of total Np present as the species NpO_2^+ is presumed to play an important role in the sorption behavior since experiments show that NpO_2^+ sorbs whereas the carbonate and hydroxy complexes do not sorb. Figure 7-103 shows the model calculations of NpO_2^+ fraction versus pH for J-13 fluids at various temperatures, and UE-25 p#1 fluid at 25° C. For the UE-25 p#1 fluid (higher carbonate concentrations) at 25°C, the shift to species other than NpO_2^+ occurs approximately one-half of a pH unit lower than for J-13 fluid. Potentially larger effects are predicted for the temperature dependence, which shifts the predicted curves up to two pH units from 25 to 90° C. This effect is due to the temperature dependence of the equilibrium constant of the mono-carbonate complex reaction (the sixth reaction in Table 7-6). If the temperature-dependence of this reaction is properly captured in the equilibrium constant values in the table, and if the temperatures in the zeolites are elevated during the period in which Np is travelling through the zeolites, then the temperature dependence will have a significant impact on performance predictions.

Sorption Data - A comprehensive set of laboratory studies have been performed to characterize the sorption of Np on Yucca Mountain tuffs (Thomas 1987; Bertetti et al. 1996; Tait et al. 1996; Triay et al. 1996a, b). In the first phase of experiments, sorption was shown to be minimal ($K_d <$

0.4 ml/g) on the vitric and devitrified tuffs in the pathways from the repository to the water table. Small but appreciable sorption (K_d values of 2.5 ± 1.4 ml/g for 46 batch sorption measurements in J-13 water in the pH range 8.25 ± 0.5 reported by Triay et al. 1996c) was found on rocks that have undergone zeolitic alteration. The presence of minor amounts of highly sorptive minerals such as hematite does not appear to translate to higher sorption in the whole rock samples. Triay et al. (1996a) postulate that this lack of enhanced sorption compared to single mineral sorption measurements could be due to passivation of the mineral surface. In fractures, coating minerals such as calcite, iron oxides, and manganese oxides exhibit significant sorption of Np (Triay et al. 1996a). However, in these simulations, we assume that matrix flow predominates in the nonwelded to partially welded units below the potential repository for the infiltration rates chosen. Therefore, sorption on fracture surfaces is not considered, although under different sets of hydrologic conditions, fracture flow and sorption on fracture surfaces could become important.

The mineral responsible for the sorption of Np on zeolitic tuffs at Yucca Mountain is clinoptilolite. Triay et al. (1996b) showed that the sorption behavior of zeolitic Yucca Mountain tuff was essentially equivalent to that of pure clinoptilolite, when the experiments are carried out under similar chemical conditions, and that differences can be understood quantitatively on the basis of the relative surface areas of the samples.

Sorption Models - To develop a quantitative geochemical model of Np sorption on zeolitic tuffs as a function of groundwater composition, we rely on the batch sorption data cited previously (Triay et al. 1996c), as well as directed laboratory studies examining the impact of fluid chemistry (Tait et al. 1996, Triay et al. 1996a). The models developed in the present study are used to simulate the sorption of NpO_2^+, where the aqueous concentration of NpO_2^+ is computed via the speciation reactions in Table 7-6. Recall that Figure 7-103 illustrated the effects of the speciation calculations in a plot of the fraction of Np present as a function of fluid composition and temperature. The figure shows that as pH increases, the fraction of NpO_2^+ decreases, resulting in a lower effective sorption coefficient K_d. An estimate of the effective sorption coefficient $K_{d,\,eff}$ in the zeolitic Calico hills units is given by $K_{d,\,eff} = f_{NpO2}\, K_{d,\,max}$, where $K_{d,\,max}$ is the maximum distribution coefficient in a system consisting of 100 percent neptunyl cation, and f_{NpO2} is the fraction of total Np present as neptunyl cation. For the J-13 fluid at 25°C, the speciation results will predict a reduction in f_{NpO2} and $K_{d,\,eff}$ by a factor of 2.9 from pH 7 to pH 8.5. These chemical calculations are in good agreement with the recently published sorption data of Triay et al. (1996c) of a decrease in K_d in J-13 water by a factor of 2 from 3 ml/g to 1.5 ml/g in this pH range (Figure 4 of that report).

The transition to Np carbonate complexes occurs at a lower pH value for UE- 25 p#1 water, consistent with the low measured sorption coefficients in the fluid even at pH 7. There is some discrepancy between the speciation calculations and the observed sorption data, since the experiments show essentially no sorption even at pH 7, while the speciation calculations predict significant amounts of NpO_2^+ are present up to a pH of about 9 for UE-25 P#1fluid. Nonetheless, the trends are correct and the pH ranges over which transitions occur are within one pH unit of what is consistent with the batch sorption data. Therefore, it was determined that the reactions of Table 7-6 provide a consistent explanation for the impact of pH on the observed sorption behavior of ^{237}Np.

To simulate the sorption of NpO_2^+ cation, the speciation results presented above are coupled with an ion exchange model for NpO_2^+. Ion exchange models for clinoptilolite have been studied by many authors including Viani and Bruton (1993), Pabalan (1994), and Triay et al. (1996a). The exchange model developed in the present study is patterned after a model first proposed by Triay et al. (1996a). Figure 7-104 shows the measured sorption coefficient on pure clinoptilolite as a function of Na^+ concentration in pure sodium bicarbonate solutions (data from Tait et al. 1996). These data indicate an ion exchange mechanism with a restricted number of sorption sites, as shown by Triay et al. (1996a). The following set of ion exchange reactions are assumed in the present study:

$$NpO_2^+ + tAl\text{–}Na^+ \Leftrightarrow tAl\text{–}NpO_2^+ + Na^+ \tag{7-35}$$

with $\log_{10} K = -1.58$

$$Ca^{2+} + 2tAl\text{–}Na^+ \Leftrightarrow (2tAl)\text{–}Ca^{2+} + 2Na^+ \tag{7-36}$$

with $\log_{10} K = 1.5$,

where *tAl* represents a tetrahedral aluminum sorption site. The first reaction alone is used to model the data of Figure 7-104. The waters at Yucca Mountain contain divalent cations like Ca^{2+} that compete for the available ion exchange sites. The equilibrium constants listed in Equations 7-35 and 7-36 along with the cation exchange capacity (CEC) for neptunyl cation of 5×10^{-4} equiv/L, produce a good match to the data of Figure 7-104. The resulting K_d is 3 ml/g in J-13 water at pH 7, which is consistent with the J-13 sorption data. As shown earlier, as pH increases, the speciation reactions decrease the relative amount of in NpO_2^+ solution, resulting in lower apparent K_d's.

There is significant variability in the measured K_d values for zeolitic tuffs that is not captured with a single set of sorption model parameters. It is assumed that the range of apparent K_d values is due to different surface areas and, hence, surface sites available for sorption. This same variability is likely to be a reality in the field, since the fraction of clinoptilolite measured in field samples within the zeolitic horizons varies significantly (Vaniman and Bish 1995). We handle this variability in the field simulations by assuming a range of surface site densities, while keeping the exchange coefficients constant.

In UE-25 p#1 water, the measured K_d values are considerably lower than in J-13 water. In the model developed here, this result is reproduced as the combined effect of a greater tendency for the formation of non-sorbing Np-carbonate complexes due to the higher carbonate concentrations and the higher concentrations of competing cations for the small number of available sorption sites. The best available data for comparison are column sorption experiments, which are more accurate for measuring low apparent K_d values than are batch experiments. The values of 0.2 to 0.4 ml/g determined in column studies by Triay et al. (1996d) are somewhat higher than the value of essentially 0 predicted by the model. This discrepancy is acceptable when making repository performance predictions, since the model errs on the conservative side (i.e., no sorption). Triay et al. (1996a) suggest a surface complexation reaction (not included in the present study) that could explain the difference. One area in which the

model may yield non-conservative predictions is in fluids with low pH (<8) but high concentrations of dissolved cations such as UE-25 p#1 fluid. For example, at pH 7, the model predicts an apparent K_d of roughly 1 ml/g, whereas experiments at these conditions yield K_d values of essentially 0. Thus, we must view the model results carefully in the regime of low pH and high cation concentrations. Additional experiments carried out under carefully controlled conditions with varying cation concentrations are needed to further refine the model. Nonetheless, the current model allows us to examine the sensitivity of the system to different chemical compositions in the range measured in the field.

7.6.2.3 Results of Neptunium Transport Simulations

7.6.2.3.1 Sensitivity of Np Transport to pH

In the site scale Np transport simulations, we study the effects of infiltration rate, temperature, pH, and bicarbonate, sodium and calcium concentrations on Np migration from the repository to the water table. The effects of pH and water composition are given here. Further information on the effects of infiltration rate and temperature are given in Robinson et al. (1997, Chapter 11.8.3). In these simulations, we examine the rate at which ^{237}Np reaches the water table as a function of time. This measure of performance was selected because the radionuclide mass flow rate is a direct input to the SZ transport model that predicts the radionuclide concentrations at the accessible environment. All simulations use the competitive ion exchange reaction model with speciation discussed above.

We investigated the influence of pH on ^{237}Np sorption and transport was investigated. The reactions considered in the model result in little predicted change of pH with time, at the zeolitic layers, even during the heating phase. As recently shown by Lichtner and Seth (1996) and others, the pH is a strong function of CO_2 degassing due to heating and boiling at the heat pipe front. Rigorously modeling the geochemistry at the heat pipe front is beyond the scope of this study since the calculation would require a gas phase model of CO_2 transport. In addition, the precipitation-dissolution of minerals such as calcite and quartz would have to be considered. To approximate CO_2 degassing due to heating, we fix the concentration of CO_2 (aq) is fixed throughout the model while allowing for all other components to dilute and concentrate due to heating effects. In effect, it was assumed that the partial pressure of CO_2 (g) is fixed throughout the mountain. To examine the effect of heating on pH, we ran a simulation starting at a pH of 7 with J-13 geochemical conditions. As the waste heat dries out the near field, the pH sharply increases to about 11 at the heat pipe front and returns to ambient conditions as the repository cools to ambient conditions (Figure 7-105). This result qualitatively agrees with the 1-D model results of Lichtner and Seth (1996). The extent of the pH rise is perhaps overpredicted in our model, since the buffering by dissolved silicic acid is not included, which has a dissociation constant (pK_a) of 9.8. However, it is shown below that this simplification results in conservative model predictions since the higher the pH at the zeolites, the less sorption of ^{237}Np. Figure 7-105 shows that the pH at the top of the first zeolitic horizon, which are 150 meters away from the repository, is not affected by the drastic pH changes at the heat pipe front. Again, these model predictions agree with the Lichtner and Seth (1996) which indicate that the pH changes at the heat pipe front are primarily a near-field affect.

The effect of pH on ^{237}Np migration is more complicated than the infiltration effect (see Section 7.6.3.3.2 below), because the pH influences both the solubility and sorption behavior. As pH rises, solubility decreases (slower near field releases), but sorption also decreases due to the results of the speciation part of the transport model (more rapid migration). Figure 7-106 shows the ^{237}Np breakthrough for initial pHs of 7, 8, and 9 (the "carrier plume" model results will be discussed shortly).

The model results indicate that effect the pH has on reducing sorption predominates over the solubility effect, which slows the release rate from the repository. Therefore, the higher the pH, the higher the peak concentration and the shorter the travel time to the repository.

We also considered a case in which a "carrier plume" or "hyperalkaline plume" of high pH migrates from the repository. The drifts of proposed repository will be lined with cement and cement-water interactions could alter the chemistry of the fluid in which ^{237}Np is transported. A detailed model for the cement-water interactions are beyond the scope of the present study, but it is well known that a transient pulse of high-pH water is expected. To study the potential effect on ^{237}Np migration, the pH of the solution is fixed at the repository at a value of 10 for 5000 years. Recall that values of pH greater than 10 are unlikely due to buffering by dissolved silicic acid. After 5000 years, we assume that the cement is passivated and has no further effect on the pH. The pH perturbation results in a "carrier plume" that in these simulations allows a significant amount of ^{237}Np to traverse the zeolitic layers with no sorption.

By contrast, when we limit the pH perturbation to 500 years rather than 5000 years, the early-arriving peak at the water table is much less pronounced. Further work in quantifying the influence of cement and other manmade materials must be carried out to better assess the possibility of enhanced mobility of ^{237}Np due to a carrier plume.

7.6.2.3.2 Sensitivity of Np Transport to Chemical Composition

In contrast to the strong influence of pH on the speciation and sorption, simulations (not shown in the figure) performed over the range of bicarbonate concentrations measured in the pore fluids illustrate that bicarbonate concentration is not a significant uncertainty affecting ^{237}Np migration. Earlier, we showed that higher carbonate waters results in the formation of a higher percentage of nonsorbing carbonate complexes. Therefore, the water with a bicarbonate concentration of 300 mg/l results in slightly less sorption than the water with 50 mg/l bicarbonate concentration. However, the range of bicarbonate concentrations in UZ fluids is significantly lower than the concentration in UE-25 p#1 fluid, where the presence of carbonates is expected to significantly lower sorption. Therefore, the influence of this parameter is not critical to the model predictions.

Variability in the concentration of cations competing for sorption sites is expected to influence the transport predictions. According to the ion exchange model, sodium and calcium concentrations play an important role in the degree of ^{237}Np sorption. However, as with bicarbonate concentration, the cation concentrations are fairly tightly constrained from the field measurements. In fact, as long as the concentrations do not evolve over time, transport predictions for the cation concentration ranges shown in Table 7-4 do not differ significantly from the results already presented. However, field evidence and climate studies indicate that infiltration rate and concentrations of dissolved ions are expected to evolve over time. Earlier it

was shown that the higher infiltration rate scenarios would result in a more rapid release of ^{237}Np, but it was assumed in those simulations that sodium and calcium concentrations were independent of infiltration rate. A transient simulation is now presented in which it is assumed that after 1000 years at 1 mm/y, a wetter climate prevails, and infiltration rate increases to 4 mm/y. In addition, in response to lower evapotranspiration rates relative to precipitation that is likely to prevail under a wetter climate, it is assumed that sodium and calcium concentrations more dilute by a factor of four during the higher infiltration period. Figure 7-107 shows that the changing climate scenario behaves more like the 1 mm/y case than the 4 mm/y case, despite the fact that the 4 mm/y infiltration rate is present during the majority of the time of ^{237}Np migration. The reason for this behavior is that ^{237}Np effective K_d increases from about 2 ml/g to 5.5 ml/g at the more dilute sodium and calcium concentrations. Therefore, if higher infiltration rates are accompanied by more dilute percolating fluids, the transport predictions will underestimate sorption if a constant K_d is assumed.

7.6.2.3.3 Sensitivity of Np Transport to Repository Heating

The influence of repository heat on the flow field, transport properties, and ^{237}Np migration may be broken into two categories. First, changes in ^{237}Np transport due to the direct influence of variable temperature and changes in the flow field in response to the temperature field. Second, changes in ^{237}Np transport due to the alteration of the geochemical and hydrogeologic characteristics of the system resulting from changes in geochemical equilibrium and mineral dissolution/precipitation. The present analysis addresses the first issue but not the second. Recall that Figure 7-104 showed that speciation and sorption could be strongly affected by temperature effects. Furthermore, the flow field itself is different than that of the ambient system. Despite these potential complications, the direct effects of repository heating on radionuclide transport is expected to be small. This is because most the radionuclide releases from the potential repository are expected to occur at times long after the thermal pulse has substantially abated. Figures 3-14 and 3-15 from Chapter 3 suggest that the temperatures in the rock will be below 40 degrees C by 20,000 to 30,000 years. This temperature is in the range of ambient, unperturbed temperature variations, which rise to about 32 degrees C at the water table (Bodvarsson et al. 1997, 11-19 and 11-20). Radionuclide releases from the potential repository are expected to occur over very much longer time frames. In Chapter 6, Section 6.6, releases of Tc are found to be low until about 20,000 years and continue for more than 100,000 years. In addition releases of Np are found to be delayed until about 20,000 years and continue for more than 1,000,000 years. Therefore, we expect that the thermal effects from waste heat will have little direct influence on radionuclide transport. However, the question of altered mineralogy and hydrogeologic properties resulting from thermal-hydrologic-chemical interactions, which could have a longer-term effect on radionuclide transport in the UZ, requires additional investigation.

7.6.2.4 Interpretations

The need to develop a reactive transport model for ^{237}Np migration in the UZ is motivated by its relatively weak (but significant) sorption, its high solubility, and long half-life. The Yucca Mountain Project has therefore expended considerable effort into characterizing the chemical properties of ^{237}Np in Yucca Mountain fluids, including solubility, speciation, and sorption. This study places these measurements into perspective for the Yucca Mountain site by combining the

chemical information with the project's latest site characterization information on the hydrology, geochemistry, and projected thermal behavior of the UZ.

The chemistry of the UZ fluids has an important influence on sorption of ^{237}Np in the zeolitic horizons, which in turn controls the transport prediction. For example, the variance in effective K_d from 0 to 2.5 ml/g translates into an order of magnitude difference in travel time through the zeolitic rocks. These travel times within the UZ are predicted to be greater than 10,000 years (one proposed regulatory time frame) for most of the ^{237}Np released. If the performance period is extended to 100,000 years or longer, ^{237}Np is predicted to reach the accessible environment in significant quantities. Thus, sorption in the zeolitic tuffs has a significant impact on predicted performance even if it is not strong enough to completely retard the migration. As expected, the percolation flux also plays a dominant role in the predicted performance, in a manner analogous to the effective K_d.

The measured variability in the chemistry of the UZ fluids, though somewhat significant, does not appear to create conditions under which effective K_d values are reduced to 0, except at pH values approaching 9. Most fluid samples have measured pH values closer to 8, creating a chemical regime in which significant sorption is predicted. Of course, bypassing of the zeolitic rock through fractures would minimize sorptive retardation; this effect is discussed further below. With regard to pH, alkaline fluid emanating from the repository region could possibly reduce sorption in the zeolites; however, for this to occur, the plume of high-pH fluid must persist to the zeolites without being buffered by the UZ fluids. Initial simulations indicate that these effects are unlikely to impact ^{237}Np transport significantly unless the high-pH carrier plume lasts for 5000 years. Further work is required to characterize and model this possibility. The competing cations in the ion exchange model appear to be constrained enough to bound the effective K_d to values of order 2 ml/g. Though there is apt to be a range of effective K_d values due to variations in zeolite abundances and groundwater composition, it is unlikely that cation concentrations will achieve a level for which K_d values drop to lower than about 1 ml/g. Finally, the bicarbonate concentrations in UZ fluids are not high enough to result in significant carbonate complexation, as is implied by the low measured sorption in batch experiments with UE-25 p#1 fluids.

Although the ion exchange sorption model with speciation reverts to an effective K_d model under constant chemical conditions, its development was critical to examine the effect of transient hydrologic and chemical conditions brought on by repository heat effects and climate change. Chemical changes that are expected to occur during the period of ^{237}Np migration may potentially affect the sorption, creating a situation that cannot be characterized with a single K_d value. In addition to pH changes due to repository materials such as cement, climate changes are expected to result in chemical changes that in turn require the full reactive transport model, rather than a simplified K_d approach. However, changes in sorption that may accompany higher infiltration during future climates are found to be conservatively bounded by the K_d model.

The main thermal-hydrologic effects are expected during the first 20,000 to 30,000 years, while this is only a small part of the expected release period for radionuclides from the engineered barrier system. Therefore, we expect that thermal-hydrologic effects will have a minimal influence on radionuclide transport in the UZ. However, thermal-hydrologic/chemical

effects may result in longer-term changes to mineralogy and hydrogeologic properties. The effects of these changes still require further analysis.

7.6.3 Episodic Transient Flow And Radionuclide Transport; Process-Level Model

(Note: much of this section can be found in Robinson et al. 1997, Chapter 6, Section 12). Both the UZ flow model (Bodvarsson et al. 1997) and the FEHM UZ transport model used in the TSPA-VA operate on the assumption that episodic transients, which are precipitation events that occur on the scale of days, are damped in the PTn and that the flux leaving the PTn and entering the TSw is constant over time. This does not include large transients that occur over the scale of thousands or tens of thousands of years. The occurrence of bomb-pulse at depth in the PTn and the naturally fractured system highlights the question whether episodic events are actually damped. Therefore, in addition to the studies which use steady state flow fields, the migration of solutes from the surface to depth under transient cases is also examined.

The exact nature of transient infiltration is not known at Yucca Mountain. However, Flint (personal communication) has suggested that infiltration only occurs every several years under extensively wet conditions and over short time frames. Therefore, to effectively test the extreme case, we implemented a set of transient simulations to examine whether very large pulses of water occurring only every 5, 8 or 10 years can create sustained fracture flow in the PTn when steady-state simulations with the same annual average infiltration rate do not. The implications are that, if the flow entering the potential repository is not uniform, then higher fluxes over shorter time periods will need to be considered both by engineering and by radionuclide transport modeling.

FEHM is used to compute both the transient unsaturated flow field and the transport in that flow field. In this application of FEHM, use the dual-permeability flow and transport model with particle tracking. Unlike other investigations of episodic flow effects which used only a single pulse and simulated to a maximum of 50 years (Bodvarsson et al. 1996, 1997), a recurring episodic pulse and simulate was used for up to thousands of years. To compare with the transport on steady state flow fields, take all the water that would be associated with a given period of time and apply it to the system over a specified short episode. Table 7-7 gives the transient model names and the parameters governing the amount of water, how often it is applied, and over what duration it is applied. For example, a 10-5-90 simulation indicates applying all of the water associated with 5 years at an average of 10 mm/yr infiltration (i.e., 50 mm) over a 90-day period once every 5 years.

7.6.3.1 Process Model Calculations of Episodic Transient Flow and Transport; Parameter Sets, Hydrogeologic Environments, and Infiltration Rates Investigated

Figure 7-108 shows the unit definitions of the three columns used in this study. The primary factors controlling the solute transport rate through these systems are (1) the infiltration rate, (2) the material properties of the PTn, and (3) the thickness of the PTn. A range of infiltration rates and multiple property sets are used in the sensitivity simulations. The PTn thickness remains fixed because the columns are extracted from the 3-D site-scale geologic model. In each of the column descriptions below, the PTn thickness is listed.

The systematic modeling approach taken for each of the three columns serves to investigate process sensitivity to parameter variations and flux variations. For each parameter set investigated, simulations of flow and transport are run with mean infiltration rates ranging between 1 and 50 mm/yr. The parameter sets we look at include: the base and modified set, 6541a and 6541b respectively; the base-case set, 6412b, as reported by Bodvarsson et al. (1997), and a fault-zone set, 6541flt, that is identical to the 6541a set, but with fracture permeability in the PTn increased by a factor of ten. The basis for increasing the permeability in the PTn as a result of tectonic activity is discussed in detail in Robinson et al. (1996). The principle behind this parameter set lies with the fact that nonwelded units tend to have very low bulk fracture permeability relative to the welded units. In fault zones, however, increased fracture permeability in the PTn occurs either as a result of more fractures or greater fracture apertures. The increased permeability in faults through the PTn has been inferred from the field air permeability testing (Rousseau et al. 1996). Indications are that the permeability in a fault zone in the nonwelded rock may increase by one to two orders of magnitude.

ESF Station 35 - Station 35 is a location where a previous 3-D simulation did not predict significant early arrivals (Robinson 1997, Chapter 6). It is also a location near which both bomb-pulse $^{36}Cl/Cl$ ratios and ratios indicative of 10,000+ year old water have been found by Fabryka-Martin et al. (1996b). The bomb-pulse signal has been correlated with structural features by Levy et al. (1997). The infiltration rate above Station 35, according to Flint et al. (1996) is about 3 mm/year. This is also a location at which the consistency between fault-zone and base-case model results was demonstrated, as compared with field data and infiltration estimates (Robinson et al. 1996). The location of the Station 35 column in Nevada State Plane (NSP) coordinates is (171277, 233393) and the PTn thickness is 42 meters.

Repository Column - The repository column was chosen to be located in a zone of high estimated infiltration in the middle of the proposed repository block. The estimated infiltration rate here is about 13 mm/yr. One reason this column was chosen is to test the implication that episodic transients penetrating the PTn are most important where the flow path reaches the potential repository horizon. The PTn is 58 meters thick here, significantly greater than over the ESF. The location of the repository column in NSP coordinates is (170600, 234000).

ESF Station 57 - Station 57 represents a unique location both in terms of the geology and the measurements that were made on samples from that location. The geology above station 57 is characterized by thinner PTn than is found further north and west (c.f. Rautman and Engstrom 1996). This also near where the Ghost Dance fault crosses the ESF. However, unlike Station 35, where bomb-pulse ^{36}Cl /Cl levels were found and attributed to a fault, no unambiguous bomb-pulse signal has been found at or near this ESF station. However, one isolated peak signal at Station 56 +93 may be suggestive of bomb-pulse. Station 57 poses high potential for bomb-pulse signal in that the PTn is only 30 meters thick.

But, the infiltration rates estimated by Flint et al. (1996) are significantly lower (about 1 mm/yr) than above other ESF stations where bomb-pulse $^{36}Cl/Cl$ has been found. The Station 57 column is located at NSP coordinates (171160, 231200)

7.6.3.2 Steady-State Results

Steady-State Flow with Parameter Set 6541a - Starting with the parameter set, 6541a (Bodvarsson et al. 1997), and different flow rates, we examine the sensitivity of transport from the surface to the ESF or potential repository horizon. Figure 7-109 shows the breakthrough curves for the three columns for flow rates ranging between 0.5 and 10 mm/yr. With these simulations, the relationship between PTn thickness, flow rate, and travel time is demonstrated. For any given flow rate, the travel times are greatest in the repository column because it has the thickest PTn. However, the difference in breakthrough curves for the three columns for the 50 percent breakthrough (the median age), is fairly small. For example, at 5 mm/yr the difference between the high and low 50 percent breakthrough is less than 2000 years. The flow rate, regardless of station location, has a significant impact on simulated travel times. For flow rates spanning the range from 0.5 to 20 mm/yr, the 50 percent arrival ranges from 20,000 down to 800 years. With $^{36}Cl/Cl$ analyses in the ESF possibly indicating ages in excess of 10,000 years, these simulations indicate that a flow rate less than 5 mm/yr is necessary at that location in order to obtain travel times on that order.

Station 57, with the thinnest PTn of the three columns, yields the fastest travel times for any given flow rate. With this parameter set, Station 57 is the only one of the three columns that predicts appreciable bomb-pulse signals, although there is a trace of bomb-pulse at the other two locations for the higher flow rates. To yield a bomb-pulse signal, sustained fracture flow in the PTn is required because simulated travel times in the matrix with these properties are always greater than 50 years, even in the thin PTn at Station 57. Sustained fracture flow in the PTn is more likely where the PTn is thinner because there is less total thickness over which water starting out in PTn fractures (being fed by TCw fractures) can be imbibed into the matrix. To demonstrate this point, Figure 7-110 shows the simulated distribution of flux between matrix and fractures in the Station 35 column and Figure 7-111 shows the simulated flux distribution in the Station 57 column. At Station 35, there are over 25 meters in the PTn (between elevation 1220 and 1250) in which the fractures are dry. Conversely, at Station 57, the PTn fracture flow only approaches zero for a very short distance. In fact, the fractures in the PTn never completely dry out and account for as much as 1 percent of the total flow at higher infiltration rates as is shown in the expanded view in Figure 7-111. This small component of high velocity flow leads to the small component of early arrival at Station 57 as shown in Figure 7-109.

In the southern portion of the study area, the PTn is thinner and simulations with the 6541a parameter set (Bodvarsson et al. 1997) lead to some early arrival at the potential repository horizon. Moving further north, the PTn thickens, and fracture flow in the PTn is no longer sustained for the entire thickness, except at isolated locations as discussed later under fault zones. These results are consistent with both the two- and 3-D simulations reported in Robinson (1997, Chapter 6).

The $^{36}Cl/Cl$ data analyses in the southern ESF have shown no bomb-pulse ratios. Yet, these simulations indicate that the southern ESF is the most likely place to find such signals. However, the infiltration rates estimated by Flint et al. (1996) are relatively lower here than further north. Thus, the findings of Robinson et al. (1996) and Fabryka-Martin et al. (1996b) which indicate that a sufficient condition for fast path travel to the ESF requires both the potential for sustained fracture flow in the PTn and adequate flow rates to yield such sustained

flow would indicate that adequate infiltration rates are really not occurring above the southern ESF. Even with this property set, the potential for sustained flow decreases with decreasing infiltration rates. Therefore, these results indicate that the infiltration rate in the region above Station 57 are low, probably lower than 1 mm/yr. Later in this section we investigate the potential for episodic infiltration events to yield fast paths to the ESF at all three of the locations considered here.

Comparing Parameter Sets - The most recent LBNL Site-Scale flow model (Bodvarsson et al. 1997) contains numerous hydrologic parameter sets that are presented as fits to the existing matrix saturation data (and to some extent, matrix potential data). In this study, the primary focus is on parameter set 6541a and variations on this data set. However, the primary parameter in the LBNL report appears to be the 6412a and 6412b sets found in Table 6-4-1.2 of that report. These sets have been compared in Robinson (1997, Chapter 5) with respect to simulated saturation profiles and simulated radionuclide transport from the potential repository to the water table. Here the differences are briefly examined simulations of ^{36}Cl from ground surface to the potential repository horizon using these two parameter sets. Figure 7-112 shows the breakthrough curve at ESF station 35 for property set 6412b. Comparing this plot with the Station 35 breakthrough in Figure 7-109 where parameter set 6541a was used, it is seen that there are substantially more early arrivals resulting from fracture flow in the PTn. To demonstrate this, Figure 7-112 also displays the flux distribution for the 5 mm/year simulation. In the PTn, using the 6541a properties, the fractures account for the majority of the flow, thus the large percent of early arrival times simulated with the breakthrough analysis. Due to the large amount of flow yielding fast travel times from the surface to the ESF, one would expect indicators of young water in both the ESF as well as the perched water if the 6412b parameter set represented the actual system. In the ESF, one would expect a ubiquitous distribution of bomb-pulse signals. Since that does not appear to be the case, and because the perched water analyses indicate water with ages between 3,000 and 15,000 years, this parameter set can be reasonably ruled out for further consideration. The implications are that a uniform scaling of the reduction in fracture-matrix coupling is probably not the best approach for calibration of material properties to matrix saturations. In this regard, the 6541 data set gives results that are in better (qualitative) agreement with observations.

Parameter Set 6541b—A Slight Modification to the 6541a Parameter Set - The 6541b parameter set is our modification to property set 6541a set in which we simply eliminate the fracture saturation based reduction in fracture-matrix coupling in the PTn. The result is virtually identical matrix saturation simulations. Figure 7-113 shows the minor difference in simulated saturations at the repository column for these two different parameter sets. There is a maximum difference in saturation of 0.3 in the upper PTn. However, the effect of this property modification on transport simulations is more significant than it is on saturation simulation, as is demonstrated in the next section.

Fault Zone PTn Properties - In zones of tectonic activity, it is highly likely that the fracture permeability of the PTn increases. The modification of PTn fracture properties in the dual permeability model is discussed in detail in Robinson et al. (1996). Here we implement a representative case from the variety of modifications investigated previously. To represent the increased fracturing in fault zones, a new property set, 6541flt, is developed. It is identical to 6541a, except the fracture permeability in the PTn is increased by one order of magnitude. The

result of this modification is such that fractures in the PTn that start off with flow are more likely to continue flowing to the bottom of the PTn because the residence time is lower with less opportunity imbibition into the matrix.

Steady Flow - Flow and transport are first simulated using FEHM (with particle tracking for transport) on each column with uniform annual infiltration rates of 1, 5, 10, 20, and 50 mm/yr. With these steady-state flow fields, particles are inserted into the fractures of the TCw and their breakthrough at the potential repository horizon is monitored. The simulated saturation and flux distribution between fractures and matrix for each flow rate and parameter set is recorded; some are plotted in this section for comparison.

Transient Flow - For the following comparisons, both steady state and transient flow are considered. The case of 10-5-10 (cumulative volume of 10 mm/yr applied once every 5 years for 10 days) is primarily considered due to its sharp contrast with steady state 10 mm/yr flow. We perform the transient flow and transport calculations using FEHM with particle tracking as discussed in Section 7.4.2.

7.6.3.3 Comparisons of Transient and Steady-State Flow and Transport; Three Property Sets for Steady-State and Transient Flow.

At each of the three stations considered in this section, the arrival times at the potential repository horizon are compared for three property sets (6541a, 6541b, and 6541flt) and for steady-state flow at 10 mm/yr and transient flow with 10-5-10 application. Figure 7-114 compares the breakthrough curves at Station 35. The fault-zone properties lead to greatest amount of early arrival. The 6541a and 6541b sets yield similar results with 6541a leading to slightly faster early arrivals. This pattern is exactly what should be expected as the potential for PTn fracture flow is decreased from 6541flt to 6541a to 6541b. The transient pulses lead to slightly more sustained flow in the fractures and thus a greater amount of early arrival, particularly for the 6541flt properties.

Figure 7-115 shows the responses at Station 57. Again the fault-zone properties lead to the most early arrival (bomb-pulse era signal), followed by the 6541a set and finally the 6541b set. Here, however, the difference between 6541a and 6541b is more substantial than at Station 35. Whereas the 6541b case does not lead to a bomb-pulse signal, the 6541a case does. It is also interesting to note that the steady-state flow fields for 6541flt and 6541a lead to more early arrival than the pulsed, episodic cases. A complete testing of all possible episodic schedules has not been conducted, but, from these limited number of runs, it is inferred what simulated processes are controlling the calculated results. In the episodic case, a large pulse of water enters the PTn fractures carrying the particles that were released at the surface. As the pulse moves through the PTn, more and more water imbibes into the matrix bringing particles into the slow moving matrix flow system as well. Without the fluid associated with steady state flow to keep the particles moving along in the fracture, more particles follow the flow into the matrix during the 5 year imbibition period. For this particular case, there is actually more imbibition in the transient flow scenario than in the steady-state flow scenario. At lower average annual infiltration rates, the trend is probably reduced because a greater percentage of the steady-state flow will imbibe into the drier PTn matrix.

Figure 7-116 shows the simulated breakthrough for the 10 mm/yr comparison at the repository column. This location has the thickest PTn and hence shows the least likelihood for sustained transient flow in the PTn. However, for the fault-zone properties there is some and the 6541a properties show slightly more fracture flow than 6541b, especially for episodic flow conditions.

Additional Transient Cases - Due to the relevance of these studies at the potential repository, the remaining simulations of transient effects are performed on the repository column only. Figure 7-117 compares the effects on early breakthrough of increasing the amount of water applied for 10 days every 5 years. Other than moving the mean breakthrough time up due to the greater total flux of water, the effect on early arrival time is not particularly different than it was for the 10 mm/yr average. This indicates a favorable damping capacity of the PTn, at least where it is thicker than near Station 57 where such pulses did not appear to be damped out.

When considering the effect of transients on facilitating fracture flow in the PTn, the duration of the transient should be considered. In Figure 7-118, we examine the difference between 10 day infiltration events and 90 day infiltration events. In each case, the average infiltration rate is 10 mm/yr, and the frequency of events is every 5 years.

Although the percent of water coming through the PTn fractures (arrivals significantly earlier than the mean travel time) is small, it is interesting that the 90 day transients apparently lead to more fracture flow in the PTn. This indicates that, for the properties uses, a greater amount of a large short pulse can be imbibed into the PTn matrix than a pulse of longer duration but equal volume. Due to the transient, this simulated behavior may shed some light on the rate at which the imbibition process occurs relative to the flow rate and frequency of flow events.

Another way to examine the effect of transients at the potential repository is to simulate the flux response to infiltration transients at the surface. Figure 7-119 shows the different responses for the three different property sets considered in this section when a 10-5-10 model of transient infiltration is specified. Whereas the 6541b properties almost completely damp any fluctuations, the 6541a properties lead to an interesting long-time fluctuation at the potential repository. The 6541flt properties allow an increase in flux of about ten percent at the potential repository after each infiltration event. The flux profile also follows the long-time variation of 6541a.

The reason for this behavior is not immediately known. Further investigations will determine if it is a response representative of the wetting and drying of PTn matrix material or just a boundary effect of the simulation. However, if it were a boundary effect, one would expect the 6541b run to behave in a similar manner.

7.6.3.4 Interpretations

1-D models are used to examine, in detail, the effects of episodic transients, fault zone properties, and the relationship between material properties, infiltration rate, and travel time from the surface to the potential repository horizon. Transient flow tended to yield different, but not always worse performance from the steady flow scenarios. The effects of these differences tend to be reduced as PTn thickness increases. However, the study's fault zone property analyses

indicate that if the PTn fracture permeability is increased due to tectonic processes, then transient effects are more likely to propagate to depth.

7.6.4 Fine-Scale Mineralogical Heterogeneity and Radionuclide Transport

(Note: much of this section can be found in Robinson et al. 1997, Chapter 12). Natural spatial variability in hydrologic and chemical properties can play an important role in the transport of contaminants in groundwater. Stochastic techniques to study the effects of this small-scale variability make up a large part of the groundwater literature (Gelhar 1993). For example, reduction in sorption due to physical and chemical heterogeneity has been described by Tompson et al. (1996) and Smith et al. (1996). They suggest that for specific saturated flow systems, significant contaminant transport will occur in zones of minimal reactivity since permeability and reactivity were found to be inversely correlated. Since the study is concerned with zeolite heterogeneity in the UZ, the relationship between reactivity and effective permeability must be considered rather than the relationship between saturated permeability and reactivity.

The site-scale model of Yucca Mountain relies on averaging techniques in order to model the large scales which are of interest in the flow and transport calculations. In this section, we investigate the effects of small-scale variability in the zeolite distribution on the transport of ^{237}Np through the UZ. We develop the methodology for generating conditional simulations of zeolitic abundance to determine whether small-scale heterogeneities will significantly affect the travel times of ^{237}Np through the UZ.

7.6.4.1 Artifacts of a Bimodal (Percent Zeolite Threshold) Representation in a Unsaturated Zone Transport Model

7.6.4.1.1 Spatial Continuity of Zeolitized Regions

Another assumption made in previous transport calculations was to use a threshold cutoff for zeolitic abundance (Robinson et al. 1997). The threshold provided a method for assigning either vitric or zeolitic properties to each node, since correlations did not exist between the degree of zeolitization and the hydrologic and chemical properties. In this section we will replace the zeolitic threshold method with more rigorous geostatistical modeling. This will allow us to determine whether using a threshold cutoff for zeolitic abundance is an appropriate upscaling method for characterizing small scale heterogeneity. Figure 7-120a shows the zeolitic distribution when a 10 percent threshold is used as the basis for considering a node to be zeolitically altered. A potential problem with using a simple threshold is that the zeolitic distributions change significantly as a function of the threshold. Figure 7-120b shows the zeolitic distribution for a 20 percent threshold: the layers loose their continuity and in some places appear as lenses. In this chapter, we examine the assumption of using a cutoff for zeolitic abundance by comparing those results to geostatistical simulations in which permeability, characteristic curve parameters for relative permeability, and reactivity are correlated to zeolitic abundance. With such correlations, the effects of chemical and physical heterogeneity can be treated more rigorously without having to rely on a threshold cutoff for zeolitic abundance.

7.6.4.1.2 Effects on Flow and Transport Patterns

We carried out example calculations for two simple systems having layered and lensed zeolitic tuff embedded in vitric tuff (Figure 7-121a). The properties of the zeolitic tuff differ from the vitric tuff in terms of saturated and effective hydraulic conductivity. The properties are given in Table 7-8.

^{237}Np is assumed to linearly sorb on the zeolitic tuff with a K_d of 2 ml/g and to not sorb on the vitric tuff. The critical feature of the property variations is that even though the vitric tuff has a much larger saturated hydraulic conductivity, the effective hydraulic conductivity of the zeolitic tuff exceeds that of the vitric tuff when the system capillary pressure (head) exceeds approximately 100m. Capillary pressures of 100m or more occur in this system for flow rates below 1mm/yr. Contact of ^{237}Np with the zeolitic tuff is independent of flow rate in the layered case because all the flow must pass through the zeolitic layer. For the lensed case, however, contact of ^{237}Np is sensitive to flow rate. At higher flow rates, flow (and hence dissolved ^{237}Np) in the lensed case preferentially by-passes the zeolitic tuff . Again, this is because the vitric tuff has a higher effective hydraulic conductivity than the zeolitic tuff at a 4 mm/yr flow rate. For flow rates of 1 mm/yr or less, the higher effective conductivity of the zeolitic rock results in greater fractions of the total flow moving through the zeolitic lens. At flow rates below 1 mm/yr, a large fraction of the total flow passes through the zeolitic lens, resulting in similar degree of contact between ^{237}Np and zeolites as for the layered case. The enhanced contact with zeolitic tuff for low flow rates in the lensed case results in ^{237}Np flux profiles, shown in Figure 7-121b, being similar to the layered case. Likewise, the bypassing of the zeolitic tuff under conditions of higher flow makes the ^{237}Np flux profile much larger at early times as compared with the layered case.

7.6.4.2 Development of a Geostatistical Mineralogical Model for Zeolite Abundance

Geostatistical techniques are powerful tools that can be applied to the Yucca Mountain mineralogic model because they (1) provide insight into the variability of the data; (2) characterize the uncertainty surrounding the data; and (3) help predict data values in unsampled areas. By utilizing careful geostatistical analysis of the 3-D data one can produce a fully populated stochastic site-scale numerical mineralogic model that can then be used as a basis for transport calculations.

The mineralogic model developed with STRATAMODEL (Robinson et al. 1997, Chapter 2) interfaces to a compatible commercially available geostatistical software package (RC^2). The regularly-spaced mineralogic data and stratigraphic surfaces output from SGM are imported by RC^2 and then analyzed by constructing experimental horizontal and vertical variograms of mineral abundance. These variograms help define the spatial continuity through a quantitative description of how data values change as a function of distance and direction. Interpretive variogram models are then used in the process of performing ordinary kriging and sequential Gaussian simulations. Ordinary kriging provides a weighted linear estimate of values at unsampled areas and is generally a smoothing technique that results in a map of the uncertainty in the data (kriging variance). Sequential Gaussian simulations, on the other hand, produce conditional simulations, given an initial random number seed. Changing the seed for each simulation yields a different realization, resulting in realistic maps of possible data values. Each

simulation honors the data and always samples the same model variograms. Examples of the variograms, kriging maps, and sequential Gaussian simulations are shown in Section 7.6.5.2.2.

7.6.4.2.1 Correlations Between Zeolite Abundance and Hydrogeologic Properties

Figure 7-120 shows how different threshold cutoffs for zeolitic abundance can result in significantly different zeolite distributions. A particular threshold was chosen for simplicity, and because correlations did not exist between the degree of zeolitization and the hydrologic and chemical properties. Therefore, this method was used to tag each spatial location as either vitric tuff or zeolitic tuff. Note, however, that the use of a threshold is still an improvement over the method used in previous studies in which entire layers were assigned either zeolitic or vitric properties.

In reality, the percent of zeolitic alteration is a continuous property which varies from 0-100 percent. This section, examines various sources of field data to determine whether the hydrologic (e.g., permeability and characteristic curve parameters for relative permeability) and chemical properties (e.g., distribution coefficient K_d) of the tuffs can be correlated to zeolitic abundance. Such correlations are expected because the physical and chemical properties of geologic medium can often be related back to fundamental measurable properties such as grain or pore size (Smith et al 1996). Specifically, the process of zeolitic alteration acts to reduce the pore sizes of the vitric tuffs. If such correlations could be developed, the need for using a threshold cutoff would be eliminated.

The correlations derived are a combination of empirical relationships obtained from field data and theoretical relationships. Several regression relationships have been developed by various researchers within the Yucca Mountain Project. Theoretical relationships with physical bases also exist and could be used to correlate the various properties. The correlations specifically describe relationships that are used to generate realizations of multiply correlated properties (permeability, characteristic curve parameters, and K_d). In particular, developed correlations between permeability and percent clinoptilolite, porosity and permeability, permeability and reactivity (K_d), and permeability and the van Genuchen capillary scale factor, alpha. Further details on the specifics of these correlations are given by Robinson et al. (1997).

7.6.4.2.2 Geostatistical Modeling of Percent Zeolite Abundance

Before the above correlations can be used in numerical simulations, realistic distributions of zeolites must be constructed. The geostatistical package RC^2 was used in conjunction with STRATAMODEL to perform several geostatistical operations. First, the stratigraphic information and the well data were ported to RC^2 , which was used to generate horizontal and vertical variograms of zeolitic abundance. The variograms provide a measure of spatial correlation by characterizing how sample data are related with distance and direction. These variograms are necessary to perform kriging and sequential Gaussian simulation. Kitanidis (1996) states that variogram analysis cannot be performed using a set procedure; instead, creating a variogram is more of an "art". A variogram should be constructed from knowledge of the underlying processes generating the data and by customization of the available tools. The main goal of a variogram analysis is to construct a variogram that best estimates the autocorrelation structure of the underlying stochastic process. For simplicity, the horizontal

variogram was chosen to be isotropic. Specifically, the spatial correlation of zeolites in the east-west direction was chosen to be the same as the spatial correlation in the north-south direction. Rautman has shown that there are some significant differences between the east-west and north-south zeolite distributions (C. Rautman, pers. comm. 1997). In the future, directional horizontal variograms will be used in the analysis.

Further details of the variogram analyses is given in Robinson et al. (1997). We find a horizontal correlation length of 2000 m and a vertical correlation length of 16 m, based on a spherical variogram model. We develop different realizations for zeolitic abundance using a sequential Gaussian simulation method. To capture the detailed spatial variations in zeolitic abundance from a given realization, we refine the zone investigated and nodes with an average spacing of 4 – 6 m. We mapped each conditional simulation onto a refined cross section through Antler Ridge, between the Solitario and Ghost Dance faults. The result is shown in Figure 7-122.

7.6.4.2.3 Comparison of Transport Behavior for Sorbing Nuclides: Hydrogeochemical Transport Properties that are a Continuous Function of Zeolite Abundance Versus a Bimodal Zeolite Abundance Representation

In this investigation, we simply use the zeolitic abundance at each spatial location and correlate it to K_{sat} , α, and K_d rather than using a more complex linear coregionalization technique. As stated above, this assumes a 1:1 correlation between properties. As work continues, linear coregionalization will be used to distribute correlated properties to relax this assumption. In addition, other parameters such as fracture spacing may be correlated to zeolitic abundance, if appropriate.

Flow and transport calculations are performed using the dual-permeability option in FEHM. All layers above the Calico Hills unit are assigned the property values of parameter set 6541a (Bodvarrson et al. 1997). The Calico Hills and Prow Pass units are given permeabilities which correspond to zeolitic abundance using the correlation discussed in Section 7.6.5.2.1. The relationship between α and K_{sat} was used to assign values for α in the Calico Hills and Prow Pass units (see Section 7.6.5.2.1). No relationship currently exists which relates zeolitic abundance to K_d, although experiments by Triay et al (1996a, b, c, d) clearly have shown that K_d increases with zeolitic abundance. As zeolitic abundance increases, the surface area available for sorption increases resulting in a higher K_d . A simple linear relationship is used to relate K_d to zeolitic abundance. At 0 percent zeolite, it is assumed that no sorption of ^{237}Np will occur. We then choose different slopes to relate the zeolitic abundance to K_d . By running simulations at several different slopes we bracket the variability in K_d.

It was found that for most property sets, even a zeolitic K_d of 20 ml/g did not strongly retard radionuclides for the 10 percent zeolitic threshold case (Robinson et al. 1997 Chapter 8.8). However, for the conditional simulation, K_d has a large effect on the travel time. Even for the case in which K_d varies linearly with zeolitic abundance from 0 to 5 ml/g, the retardation is much larger than the retardation predicted by the K_d of 20, 10 percent cutoff case (Figure 7-123). Figure 7-124 shows that for the 1/3 infiltration rate case (discussed in previous chapters), the conditional simulation again results in much larger retardation.

The reason for larger retardation of ^{237}Np for the conditional simulation is due to small, but significant, K_d at locations with zeolite abundance less than 10 percent. The increased sorption is not caused by high K_d s of 5 ml/g at high zeolite percentages. Very little flow travels through these highly zeolitized locations since they are very low in permeability. One can prove this by returning to the 10 percent threshold case. If one assigns a small K_d (just 10 percent of the maximum K_d) to all locations with zeolite abundance between 0 and 10 percent, one proves that even small but significant amounts of sorption on rock with low zeolite abundance can strongly affect the travel times of ^{237}Np (Figure 7-125). Figure 7-125 shows that modifying the 10 percent threshold case to include some sorption for locations with zeolites abundances less than 10 percent closely matches the conditional simulation.

7.6.4.3 Interpretations

A key conclusion to this study is to recommend that experimentalists determine whether rocks with low but non-zero zeolitic abundance exhibit measurable sorption of radionuclides like ^{237}Np and Uranium. Rocks with high zeolitic abundance may not be as effective in retarding these radionuclides since these rocks generally have low permeability, and contaminants would primarily enter these regions through molecular diffusion. In addition, any fractures in these highly zeolitized rocks will provide a mechanism for fluid and radionuclides to bypass these regions. However, small but significant sorption of rocks with low zeolite content may have a very large impact on the travel times for ^{237}Np through the UZ. ^{237}Np transports through the matrix of these rocks and even a small K_d of 1 cc/g can have a large impact. For the purposes of abstracting these results, we employ the small, but nonzero K_d 's identified in the vitric tuffs (see Section 7.5.3.2).

7.7 SUMMARY AND DISCUSSION

7.7.1 Summary

TSPA-VA Approach - UZ radionuclide transport calculations are computed using the particle tracking method available in the unsaturated flow and transport code FEHM. This computational method for UZ radionuclide transport is dynamically linked with the TSPA code, RIP, for these calculations. A 3-D, dual-permeability model is used for computing radionuclide transport in a fracture/matrix system. 3-D, steady, UZ flow fields, computed with the unsaturated flow code, TOUGH2, are used for the particle tracking transport calculations. Changes in the flow field due to climate change are accounted for through a quasi-steady flow and transport approximation. The transport model includes the effects of fracture/matrix interaction driven by advective and diffusive exchange between the fracture and matrix continua. The transport model includes the effects of radionuclide sorption in the rock matrix using a bounding, minimum K_d modeling approach. Colloids are included as a mechanism for the transport of plutonium. The model for aqueous plutonium transport allows for reversible sorption of plutonium on colloids. Plutonium releases from the engineered barrier system also include a fraction plutonium irreversibly sorbed to colloids.

Radionuclide mass flux at the engineered barrier system boundary is provided by RIP for the UZ radionuclide transport calculations. The radionuclide mass flux from any source within a potential repository region is distributed uniformly over the computational grids for the transport

model that lie within the region. Radionuclide mass flux at the water table from the fracture and matrix continua are mixed in a RIP cell and provided to the SZ radionuclide transport model.

Enhancements to the Particle Tracking Method for TSPA-VA - We have enhanced the particle tracking method to capture the effects of long-term transient flow and climate change and colloid-facilitated radionuclide transport.

Long-Term Transient Flow and Climate Change

We have developed an approximate method for computing UZ radionuclide transport under the long-term transient flow conditions associated with climate change. The approximation was developed due to the computational burden associated with the complete transient flow and transport calculation. The method is based on a quasi-steady flow approximation, in which the flow fields instantaneously change from one steady flow field to another in response to changing infiltration rates caused by climate change. The transport calculations restart upon change in climate using the new flow and water saturation distributions. Radionuclides present in the UZ at the time of climate change are also restarted as a distributed source of radionuclide mass in the UZ flow and transport grid. The method is shown to provide an excellent approximation to the complete unsaturated transient flow and transport problem resulting from climate change.

Colloid-Facilitated Radionuclide Transport

We have developed an approximate computational method to treat the effects of radionuclide interactions with colloids. This method is applied in the TSPA-VA calculations to the problem of plutonium transport. Previous TSPA analyses did not include the effects of colloid-facilitated radionuclide transport. The treatment addresses two types of colloidal interactions with plutonium: reversible sorption between aqueous and colloidal plutonium and plutonium irreversibly attached to colloids. For reversibly sorbed plutonium, the approximate colloid-facilitated transport method assumes linear, equilibrium partitioning of aqueous and colloidally-sorbed fractions. Colloid movement is represented by advective transport in fractures or matrix with no diffusion of colloids between fractures and matrix, and no colloid filtration.

Unsaturated Zone Radionuclide Transport Sensitivity Investigations—We have carried out additional sensitivity investigations to provide a better understanding of transport parameter sensitivity, identify the range of conditions under which a K_d sorption model may be used to bound geochemical interactions, investigate the effects of episodic transient flow on radionuclide transport, and investigate the effects of fine-scale mineralogic heterogeneity.

Parameter Sensitivity

Our sensitivity studies show that for both the 2-D flow and 3-D base-case models, radionuclide transport in the UZ is not very sensitive to variations in: matrix diffusion coefficients, fracture surface sorption, and matrix sorption individually. But the combined effects of matrix diffusion (for $D_m > 10^{-12}$ m^2/sec) and matrix sorption can significantly reduce the peak mass flow rates and delay the corresponding peak arrival time.

Increasing the radionuclide release period (given the same total mass release of radionuclides) can effectively reduce the peak mass flow rate and delay the corresponding peak mass arrival time at the water table. Such effects are far larger than the effects of matrix diffusion, matrix sorption, and fracture surface sorption on the radionuclide transport process. Thus, the performance of the Engineered Barrier System has a large effect on the performance of the unsaturated.

Different flow configurations can have a strong impact on the transport of radionuclides in the UZ. The important difference for different flow configurations appears to be the extent of matrix imbibition of flow moving through fractures. The current base-case and alternate conceptual flow models differ in the treatment of fracture/matrix interaction, primarily in the nonwelded units. However, the effects of lateral diversion and focused flow through nonwelded units in some areas of the UZ below the potential repository lead to fracture-dominated flow patterns in the nonwelded units in these areas for both the base-case and the DKM Weeps conceptual flow models. Therefore, the UZ flow conceptual model has a smaller effect on radionuclide transport through the UZ than might be expected.

Despite the relatively low fracture/matrix exchange expected in the TSw hydrogeologic units and generally lower sorption potential, this unit appears to have the largest impact for retardation of sorbing radionuclides. The main reason is believed to be a result of the flow field. The flow is relatively 1-D through the TSw. At the TSw/CHn interface, the flow becomes much more 3-D, with large-scale lateral diversion found in the northern portion of the potential repository, associated with the perched water bodies. The diverted flow is expected to find its way to the water table in a fairly concentrated flow once the flow paths have reached the edge of the perched water bodies. Therefore, a substantial amount of flow above the potential repository is focused into a relatively narrow area for passage through the CHn. This tends to diminish the effectiveness of the CHn on retarding radionuclide transport.

Sorption Models and Radionuclide Transport; an Investigation of ^{237}Np

The measured variability in the chemistry of the UZ fluids, though somewhat significant, does not appear to create conditions under which effective K_d values are reduced to 0, except at pH values approaching 9. Most fluid samples have measured pH values closer to 8, creating a chemical regime in which significant sorption is predicted. Of course, bypassing of the zeolitic rock through fractures would minimize sorptive retardation; this effect is discussed further below. With regard to pH, alkaline fluid emanating from the repository region could possibly reduce sorption in the zeolites; however, for this to occur, the plume of high-pH fluid must persist to the zeolites without being buffered by the UZ fluids. Initial simulations indicate that these effects are unlikely to impact ^{237}Np transport significantly unless the high-pH carrier plume lasts for 5000 years. Further work is required to characterize and model this possibility. The competing cations in the ion exchange model appear to be constrained enough to bound the effective K_d to values of order 2 ml/g. Though there is apt to be a range of effective K_d values due to variations in zeolite abundances and groundwater composition, it is unlikely that cation concentrations will achieve a level for which K_d values drop to lower than about 1 ml/g. Finally, the bicarbonate concentrations in UZ fluids are not high enough to result in significant carbonate complexation, as is implied by the low measured sorption in batch experiments with UE-25 p#1 fluids.

Although the ion exchange sorption model with speciation reverts to an effective K_d model under constant chemical conditions, its development was critical to examine the effect of transient hydrologic and chemical conditions brought on by repository heat effects and climate change. Chemical changes that are expected to occur during the period of ^{237}Np migration may potentially affect the sorption, creating a situation that cannot be characterized with a single K_d value. In addition to pH changes due to repository materials such as cement, climate changes are expected to result in chemical changes that can effect the transport of ^{237}Np. However, in the case of climate change, the present K_d model for sorption is expected to provide a conservative bound on transport behavior.

Episodic Transient Flow and Transport

We have used 1-D model calculations to examine, in detail, the effects of episodic transients, fault zone properties, and the relationship between material properties, infiltration rate, and travel time from the surface to the potential repository horizon. Transient flow tended to yield different, but not always worse performance from the steady flow scenarios. The effects of these differences tend to be reduced as PTn thickness increases. However, the fault zone property analyses indicate that if the PTn fracture permeability is increased due to tectonic processes, then transient effects are more likely to propagate to depth.

Fine-Scale Mineralogic Heterogeneity

We have investigated the effects of fine-scale mineralogic heterogeneity with respect to variable zeolitic alteration. Based on these studies, we have identified the importance of including small, but nonzero sorption in the Calico Hills vitric zones. The sorption data given in Section 7.5.3.2 captures these small sorption coefficients, along with their uncertainty.

7.7.2 Discussion

The results of investigations into radionuclide transport through the UZ lead to the following conclusions:

A. Under the dominant LTA climate, releases of nonsorbing or weakly sorbing radionuclides from the northern portion of the repository have short travel times, only a few years, because of fracture-dominated transport.

B. Under the dominant LTA climate, releases of nonsorbing or weakly sorbing radionuclides from the southern portion of the repository may have much longer travel times (hundreds of years).

C. For most of the mass of strongly sorbing radionuclides, such as plutonium, arrival at the water table is delayed tens to hundreds of thousands of years. Radionuclides that reversibly sorb onto colloids are also delayed; however, irreversibly bound radionuclides may behave as nonsorbing radionuclides.

D. Matrix diffusion has little effect on transport of nonsorbing radionuclides. However, combining matrix diffusion with sorption significantly retards radionuclides compared with sorption and no matrix diffusion.

E. The TSw unit is the primary barrier to radionuclide transport in the UZ because of slower fracture transport in this unit compared with the Calico Hills zeolitic unit, and its greater thickness and continuity compared with the Calico Hills vitric unit.

F. Alternative flow models like the dual-permeability model with Weeps modeling, which have smaller coupling strengths for effective fracture/matrix contact area, increase fracture-dominated transport, particularly for sorbing radionuclides. However, the magnitude of this effect is not large for weakly sorbing radionuclides.

The following items are viewed as the areas where additional work would provide the greatest improvements in the comprehensiveness and credibility of the radionuclide transport model in the UZ:

Effects of Thermal-Hydrologic-Chemical Alteration—The current model does not account for alteration of the UZ because of thermal alteration of minerals, chemical interactions of repository materials, mineral dissolution, and precipitation. These effects are potentially important to transport behavior in the UZ and need to be addressed. Thermal-chemical alteration could cause reduced matrix sorption and fracture/matrix interaction. These effects could cause increased release rates from the UZ for base case transport results; however, it is expected that our existing sensitivity studies bound most of these potentially nonconservative interactions.

Colloid Filtration—The ability of colloids to facilitate radionuclide transport is a function of their ability to migrate over large distances without being "filtered out" by the host rock. This filtration effect was not included in the TSPA because of inadequate information to bound the mechanism. The filtration effect is particularly important for the fraction of radionuclides that are irreversibly bound to colloids. However, the assumption that there is no colloid filtration is conservative for transport in the UZ.

Fracture Sorption—The effects of higher infiltration determined in this TSPA guarantee that transport will be fracture-dominated in many of the unsaturated-zone units. Minerals that line fractures are known to sorb radionuclides, but more information is needed to define the distribution and character of the fracture materials so that sorption on fracture surfaces may be included. However, the assumption that radionuclides do not sorb onto fracture surfaces is conservative for transport in the UZ. Although the present sensitivity studies have found that transport is not highly sensitive to fracture sorption, this result requires further investigation to better define the appropriate range of parameters for fracture sorption.

Matrix Diffusion—Radionuclide transport in the UZ is sensitive to changes in matrix diffusion. This sensitivity suggests that further effort is needed to bound the magnitude of matrix diffusion. In particular, the way in which the fracture/matrix contact area is used for calibrating the flow model suggests that some type of coupling strength may be appropriate for matrix diffusion. However, sensitivity studies presented in Chapter 11 suggest that the influence of matrix diffusion on total system performance is small.

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